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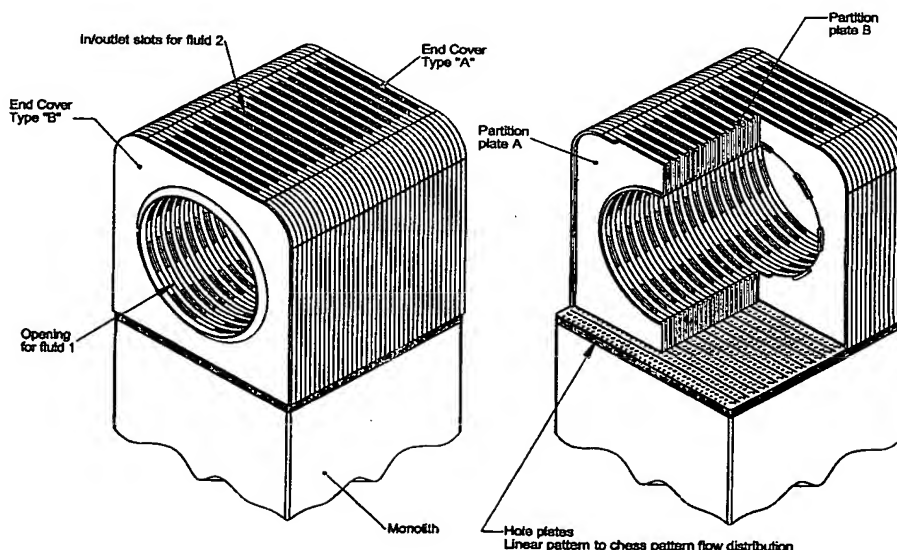
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ning of each regular issue of the PCT Gazette.

(54) Title: METHOD AND EQUIPMENT FOR DISTRIBUTION OF TWO FLUIDS INTO AND OUT OF THE CHANNELS IN
A MULTI-CHANNEL MONOLITHIC STRUCTURE AND USE THEREOF



(57) Abstract: The present invention concerns a method and equipment for distribution of two fluids into and out of the channels in a multi-channel monolithic structure (monolith) where the channel openings are spread over the entire cross-sectional area of said structure. Said equipment consists of a manifold head, a monolith unit or a monolith stack, or a row of said units or stacks or a monolith block. Furthermore, the present invention concerns a method and a reactor for mass and/or heat transfer between two fluids where said fluids are distributed through one or more of said manifold heads and units or stack or row of units or stacks or blocks.

5 **"Method and equipment for distribution of two fluids into and out of the channels in a multi-channel monolithic structure and use thereof"**

10 The present invention concerns a method and equipment for distribution of two fluids into and out of the channels in a multi-channel monolithic structure (monolith) where the channel openings are spread over the entire cross-sectional area of said structure.

15 The present invention is applicable in processes for mass and/or heat transfer between two fluids.

The two fluids will normally be two gases with different chemical and/or physical properties. But the present invention is also applicable when one fluid is a gas and the other is a liquid. One can even have systems where one or both fluids is a mixture of gas and liquid. This gas liquid mixture can constitute a continuous or homogeneous phase or a distinct two-phase flow (slug flow). In the following description the two fluids are exemplified by the name of fluid 1 and fluid 2.

25 Fluid 1 and fluid 2 are fed into said channels for fluid 1 and said channels for fluid 2 respectively. Fluid 1 and fluid 2 are distributed in the monolith in such a way that they have joint walls separating fluid 1 and fluid 2. The walls that are joint walls for the two fluids will then constitute a contact area between the two fluids that is available for mass and/or heat transfer. This means that the fluids must be fed into channels where the channel openings are spread over the entire cross-sectional area of the monolith. The present invention makes it possible to utilise the entire contact area or all of the monolith's channel walls directly for heat and/or mass transfer between fluid 1 and fluid 2. This means that the channel for one fluid will always have the other fluid on the other side of its channel walls, i.e. all adjacent or neighbouring channels for fluid 1 contain

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fluid 2 and vice versa. The present invention is particularly applicable for process intensification because monolithic structures with channel openings that have a small cross-section area (i.e. channel openings with 1-6 mm width) and thin walls can be utilized. Channels with small cross-section area and thin walls give large surface area per volume unit and thus a very compact and energy-efficient device for heat and or mass transfer.

In the present invention the contact area wall in the monolith can be a membrane capable of selectively transporting one or more components between the two fluids. Furthermore the present invention can also be utilised for two-phase flow systems where gas and liquid is transported within the same channel (here fluid 1) and perform internal mass transfer (absorption or desorption) between the two- phases (gas and liquid) simultaneously as being heated or cooled by fluid 2 through the contact area wall.

The wall between the two different fluids can also consist of active surface components on one or both sides. Such active surface components or catalysts are used when one or more chemical reactions are involved. Often chemical reactions produce or consume heat (exothermic and endothermic reactions). To optimise such reaction systems temperature control is of great importance.

A characteristic feature of multi-channel monolithic structures (monoliths) is that they consist of a body with a large number of internal longitudinal and parallel channels. The entire monolith with all its channels can be made in one operation, and the production technique used is normally extrusion.

By using extrusion technology for production of a monolithic structure, there is great opportunity to influence the geometric shape of the channels. Extrusion as a production method means that the entire monolithic structure can be made in one operation. The channels' cross-sectional area may differ in both shape and size or can be made uniform in size and shape, which is most common, for example triangular, square or hexagonal. However, combinations of several

geometric shapes are also conceivable. The geometric shape, together with the channel opening width or area, will be significant for the mechanical strength and available surface area per volume unit.

- 5 The width of the channel openings are typically in the order of 1-6 mm in size, and the wall thickness is normally 0.1-1 mm. A multi-channel monolithic structure with channel opening width of the smallest sizes stated achieves a large surface area per volume unit. The typical values for said surface area per volume unit will be in the range of 250 to 1000 m²/m³. Another advantage of
- 10 monoliths is the straight channels, which produce low flow resistance for the fluid. The monoliths are normally made of ceramic or metallic materials that tolerate high temperatures. This makes them robust and particularly applicable in high-temperature processes.
- 15 In industrial or commercial contexts, monoliths are mainly used where only one fluid flows through all the channels in the monolith. The channel walls in the monolith may be coated with a catalyst that causes a chemical reaction in the fluid flowing through. An example of this is monolithic structures in vehicle exhaust systems. The exhaust gas heats the walls in the monolith to a
- 20 temperature that causes the catalyst to activate oxidation of undesired components in the exhaust gas.

Monolithic structures are also used to transfer heat from combustion gases or exhaust gases to incoming air for combustion processes. One method involves

25 two gases, for example a hot and a cold gas, flowing alternately through the monolith. With such a method, for example, the exhaust gas can heat up the monolithic structure and subsequently emit heat to cold air. Such regenerative heat exchange processes with cycles in which there is alternation between two fluids (one hot, one cold) in the same structure is not, however, suitable where

30 mixing of the two fluids are undesirable or where stable and continuous heat and/or mass transfer is desired.

The industrial use of monoliths is limited mainly to applications in which only one fluid flows through all the channels at the same time.

5 In the literature, a number of processes or applications are described in which monoliths can be used to advantage to transfer heat and/or mass between two different fluid flows. Small-scale experimental tests have also been carried out with such processes. An example of this is production of synthesis gas (CO and H₂). Synthesis gas is normally produced using steam methane reformation. This is an endothermic reaction in which methane and steam react to form synthesis
10 gas. Such a process can be carried out in a monolith in which an exothermic reaction in adjacent channels supplies heat to the steam methane reformation.

Although it has been shown that it will be advantageous to use monoliths for mass and/or heat exchange between two fluids in a number of applications,
15 industrial use of monoliths for such applications is not very widespread. One of the most important points of complaint or reasons why monoliths are not used in this area is that the prior art technology for feeding and distributing the two fluids into and out of the monolith's separate channels is complicated and not very suitable for scaling up (i.e. interconnection of several monolith units),
20 particularly when the large number of channels in a monolith are taken into consideration.

German patent DE 196 53 989 describes a device and a method for feeding two fluids into the monolith's channels through feed pipes. These feed pipes or
25 tubes feed the two fluids into the monolith's respective channels from the plenum chambers of the respective fluids. The plenum chambers are mounted together in such a way that tubes from the outer chamber must be fed through the inner chamber and subsequently into the monolith's channels. Each individual tube must be sealed in order to prevent leakage from the channels of
30 the monolith and from lead-throughs in the walls of the plenum chambers. When heated, the monolith, plenum walls, pipes and sealing material will expand, and, when cooled, they will contract. This increases the likelihood of

crack formation and undesired leakage with mixture of the two fluids as a consequence. This likelihood will increase with the number of pipe lead-throughs.

- 5 In DE 196 53 989, the inlet and outlet zones with the sealed pipes are cooled so that a low-temperature, flexible sealing material can be used and the risk of crack formation and leakage can be reduced. A cooling system will naturally make the monolithic structure more expensive and more complicated, particularly for applications on a large scale in which the monolith consists of
10 many thousand channels and in which it is also necessary to use many monolithic structures in series and/or in parallel to achieve a sufficient surface area.

- US Patent 4271110 describes another method for feeding two fluids in and out.
15 This method has the advantage that pipe in-feeds from the plenum chamber to the channels of the respective fluids in the monolithic structure can be dispensed with completely. This is achieved by cutting parallel gaps down the ends of the monolith. These cuts or gaps lead into or out of the channels for one of the fluids. The gaps cut then correspond to a plenum chamber for the row of
20 channels that the gap cuts through. By sealing the gap's opening that faces out towards the end of the monolith, openings are created in the sidewall of the monolith where one of the fluids can enter or leave. The other fluid will then enter or leave at the short end of the monolith in the remaining open channels. A major disadvantage of this method, apart from the necessary processing
25 (cutting and sealing) of the monolithic structure itself, is that only half of the available area for mass and/or heat exchange can be utilised. For example, square channels for one fluid and the other fluid will have to lie in connected rows so that the channel structure for the two fluids corresponds to a plate heat exchanger. If the channels for the two fluids were distributed as in a chessboard
30 pattern, where the black fields correspond to channels for one fluid and the white fields correspond to channels for the other fluid, the maximum utilisation of the area could be achieved because, in such a fluid distribution pattern, all

the walls of the channels for one fluid would be joint or shared walls with those of the other fluid. With fluid channels for the same fluid in a row as in US patent 4271110, roughly only half of the channels' walls will be in contact with those of the other fluid.

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The main object of the present invention was to arrive at a method and equipment for feeding and distributing two fluids into and out of a multi-channel monolithic structure in which maximum surface area utilisation is achieved.

- 10 Another object of the invention was to arrive at an improved method and reactor for mass and/or heat transfer between two fluids.

In accordance with the invention the first object is accomplished in a method in that one fluid is fed through a slot in one or more gaps in a manifold head which
15 is sealed to one face of said monolith structure, the other fluid is fed into a tunnel in said manifold head and further through slots in said tunnel wall and into one or more gaps in said manifold head, said fluids are distributed from their respective gaps into said channels in such a way that at least one channel wall is in common for said fluids, said fluids are collected in their respectively
20 gaps in a manifold head which is sealed at the opposite side of said structure where the first manifold head is sealed, the fluids are then respectively led through a slot from one or more gaps and slots in a tunnel wall in said last mentioned manifold head.

- 25 In accordance with the invention the first object is accomplished in a manifold head in that said manifold head comprises at least three parallel dividing plates joined together with spacers to form gaps with slots between said plates and end cover plates joined in parallel to said dividing plates where said dividing plates and cover plates have one opening forming a tunnel with slots through
30 said joined plates.

In accordance with the invention the first object is accomplished in a unit in that a multi-channel monolithic structure where the channel openings are spread over the entire cross-sectional area of said structure and said channels have joint walls and said manifold head which is sealed to at least one face of said structure.

In accordance with the invention the first object is accomplished in a stack in that said stack comprises two or more multi-channel monolithic structures where the channel openings are spread over the entire cross-sectional area of said structures and said channels have joint walls, at least one of said manifold head which is sealed to at least one face of said structure, at least one plate with holes which is sealed between said manifold head and said structure on said side where the channel openings are, and at least one connector plate or other coupling device between units.

In accordance with the invention the first object is accomplished in a row in that said row comprises said units or stacks coupled together.

Typically the length of the row is in the same order of magnitude as the height of the individual stack to fit into a cylindrical shell.

In accordance with the invention the first object is accomplished in a block in that said block comprises rows of said units or stacks which are stapled face to face.

The block has the same height as the individual monolith stack, with the same width as the row and the block length proportional with the number of rows.

In accordance with the invention the second object is accomplished in a reactor in that one or more of said units or stacks or said row of units or stacks or said blocks are integrated in said reactor.

The pressure vessel contains the monolith block (the monolith structures packed closely together) with hollow spaces, ducts, channels or pipes within shell transporting one or both fluids into and out of the monolith structures as well as in and out of the pressure vessel.

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In accordance with the invention the second object is accomplished in a method in that said two fluids are distributed through one or more of said units or stacks, or row of units or stacks, or blocks.

10 Between the manifold head and the monolith one or more plates with holes for the fluids are fitted in to ensure even flow distribution and transformation of fluid flow between chessboard pattern (in monolith) and linear pattern (in manifold head).

15 The present invention makes possible to connect two or more monolithic structures through a flexible coupling integrated in the manifold head. If it is required to connect several such units together, it is essential that they can move relative to each other because of differences in thermal expansion. A number of monolith structures coupled together constitute a monolith row.

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Furthermore, the present invention makes possible to arrange a large number of monolithic structures within a pressure vessel without increasing the diameter of the pressure vessel when increasing the number of monolith structures. The system capacity can thus be decreased/increased simply by changing number
25 of rows or number of monolith structures and adjusting length of pressure vessel.

The present invention also makes possible to allow one fluid to be kept in a tubular, closed system, i.e. a pipe, and the other fluid to flow in and out from
30 hollow spaces within a pressure vessel.

If the present invention is used, it is not necessary to have cuts as described in US 4271110 or pipe in-feeds as described in DE 19653989 C2.

5 The present invention grants users the freedom to use all types of shape and size and the opportunity to utilise the maximum available surface area for heat and/or mass exchange. The method described in US 4271110 requires that all channels with the same fluid share at least one wall so that when the shared wall is removed or machined away, a connecting gap will be created that will constitute a joint plenum chamber for the fluid. The fact that two neighbouring
10 channels with the same fluid must have at least one joint channel wall means that the available heat and/or mass exchange area is reduced. In DE 19653989 C2, pipes are used that are fed from the plenum chambers of the respective fluids into the monolith channels, which can be distributed in such a way that the maximum available area can be utilised, i.e. the fluids are fed in distributed
15 in such a way that one fluid always shares or has joint channel walls with the other fluid. The two fluids are distributed in the channels corresponding to a chessboard pattern. This produces maximum utilisation of the available mass and/or heat exchange area.

20 The present invention consists of a method and equipment that can, in an efficient manner, feed and distribute two different fluids into and out of their respective channels in a multi-channel monolithic structure. It is necessary that the channel openings for the two fluids are evenly distributed or spread over the entire cross-sectional area of the monolith and that the channels have joint
25 walls. The equipment will, in an efficient, simple manner, collect the same type of fluid, for example fluid 1, from all channels containing this fluid in an inlet or outlet so that fluid 1 can be kept separate from fluid 2 and vice versa.

Moreover, the fewest possible number of parts or components and the least
30 possible processing and adaptation of these parts or components and the monolith will be favourable with regard to robustness, complexity and cost. In principle, it is true to say that the fewer individual components or parts, the

greater the advantage achieved. This contributes to simplifying the sealing between the two fluids that are to be fed into and out of the monolith's channels. The possibility of parallel fabrication of manifolds head, hole plates and the monolith structures will reduce processing time. Pre-assembling these components into a monolith unit, a monolith stack, a row of units or stacks or a monolith block will further be very advantageous for installation within a pressure vessel.

Moreover, it may be favourable to achieve the largest possible contact area (surface area) in a monolith with a given channel opening width. This will be particularly advantageous if the monolithic structure or channel walls are used as a membrane, for example hydrogen or an oxygen transporting membrane.

To achieve the largest possible transport capacity of the relevant fluid component per volume unit of the monolithic structure, it will be important to have the largest possible contact area per volume unit. It is therefore desirable for the fluid that flows in one channel to have the other fluid on all sidewalls that make up the channel. Using channels with a square cross-section as an example, the two fluids must flow through the monolith in a channel pattern corresponding to a chessboard, i.e. one fluid in "white" channels and the other fluid in "black" channels. In addition to being very significant for mass transfer between two fluids, the largest possible direct contact area will also be important for heat transfer efficiency.

The smaller the channel openings are, the larger the specific surface area in the monolith will be. To achieve compact solutions, it will therefore be desirable to have as small channels as practically possible.

At those faces of the monolith, where the monolith's channels have their inlets and outlets, a manifold head is sealed over the monolith's channel openings. For some applications, it may be necessary to seal just one face of the monolith with a manifold head. The manifold head comprises dividing plates fitted at a

distance adapted to the channel opening size in the monolith. The distance or space between the plates collects fluid from the channel openings that lie in the same row (i.e. same fluid) in the monolith. This space is called the plenum gap. In one application these dividing plates have a hole (e.g. circular hole) such that one of the fluids can be led out of or in to the tubular space formed by said dividing plates. This tubular space can be connected to a tube or pipe. Thus, if the monoliths are arranged within a pressure vessel, one of the fluids can be kept in a closed piping system connected to the tubular space of the manifold head, and the other fluid can be allowed to flow in the open space and/or via guiding ducts to the inlet and outlet openings of the manifold head in said vessel. With such a system one avoids a direct (sealed) connection to the monolith for one of the fluids.

The rows of the channel openings preferably run transversely over the entire short end of the monolith and comprise either inlet or outlet for the same fluid. These rows of fluid channel openings with the same fluid are kept separate by the sealed dividing plates in the manifold head. The two fluids will then be collected in their respective plenum gaps. With rows of channel openings for the same fluid, the plenum gap for one fluid will have the plenum gap for the other fluid on the other side of the dividing plate. In a monolith with square channels in which the same fluid is arranged in rows, the dividing plates will have to be sealed to the channel walls in the monolith. Instead of sealing the dividing plates directly to the channel walls in the monolith, one plate may alternatively first be sealed to the short face of the monolith. Said plate is a plate with holes (hole plate) through which the channel openings in the monolith lead out, i.e. so that fluid from the various channels that contain the same fluid can be fed out through the holes in said plate and into the plenum gaps. This means that the dividing plates in the manifold head are sealed to the hole plate between the rows of holes instead of directly to the monolith's channel walls that separate the two fluids.

By sealing a hole plate to one or both faces of the monolith with openings adapted for fluid 1 and fluid 2, the manifold head described can be used where the channels for fluid 1 and fluid 2 are distributed in a chessboard pattern in the monolith. This represents a method and equipment for feeding two separate fluids in and out that enable maximum utilisation of the surface area in the monolith. The fluids will be transferred from a chessboard distribution pattern in the monolith to rows of holes in the plate sealed to the monolith. Moreover, fluid 1 and fluid 2 will be fed from these rows of holes out of or into the monolith's channels where fluid 1 and fluid 2 are distributed as in a chessboard pattern with one fluid in the "black" channels and the other fluid in the "white" channels. The hole plate allows fluid distributed in a chessboard pattern to be fed out into plenum gaps divided by dividing plates that can separate fluid 1 and fluid 2 from each other. The plate's holes must have a slightly smaller opening area than the channel openings to which they are sealed. In addition to a reduced outlet area in relation to the channel area, the openings in the plate that is sealed to the monolith's channel structure and the dividing plates in the manifold head must also be designed and located so that the distance between the holes that lead into or out of the two fluids' channels is such that it is possible to place the dividing plates between the rows of holes with inlets and/or outlets for the same fluid. Using the example of square channel openings in which the two fluids are distributed as in a chessboard pattern, the dividing plates between the two fluids will follow the straight line between the rows of holes with the same fluid.

It is now possible to have two fluids distributed in channels in a monolithic structure out of or into separate plenum gaps where the channel openings are distributed in a chess board pattern. In order to be able to keep the two fluids separate when they enter or leave the plenum gaps in the manifold head, the same fluid can be fed to openings in the plenum gaps in a side edge of the manifold head, and, correspondingly, all plenum gaps for the other fluid are led out on the opposite side edge of the manifold head to the first fluid. Alternatively one of the fluids can be lead in and/ or out from the plenum gaps to a tubular space in the dividing plates and then connected or coupled to a pipe or to a

circular connection or joint to a neighbouring manifold head of a monolithic stack. Such a coupling or joint between manifold heads makes it possible to stable or arrange several monolithic units or stacks in rows. Such a row can then again be stapled close to an adjacent row. Thus the monolith units can be
5 arranged close together enabling compact solutions of multiple monolithic stacks into a monolith block or core within a pressure vessel.

In a system in which there is not only a single hole plate that feeds the fluid from each channel through the holes in said plate and directly out into the manifold
10 head's plenum gaps (the space between the dividing plates in the manifold head), but a system of two or more plates, the distance between the dividing plates in the manifold head can be made far larger than the channel openings in the monolith and thus not limited by the cross sectional area (width) of the monolith channels.

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This is done by feeding the fluid from one channel over into the flow from the neighbouring channel through channels or funnels created inside the hole plate system between the monolith and the manifold head. Fluid from one or more neighbouring channels in the monolith must then be fed out through a joint
20 outlet to the plenum gaps in the manifold head. These joint outlets/inlets are arranged in a system so that outlets for the same fluid are gathered together and, correspondingly, the outlets for the other fluid are also gathered together. These collections of outlets for the same fluid are gathered together so that they create a pattern that causes the dividing plates in the manifold head to have a
25 much greater distance from each other than if the plates were sealed directly to the manifold head, where the width of the individual channel openings in the monolith would determine the distance.

The most efficient heat transfer per volume unit of monolithic structure is
30 achieved with small channels and fluid distribution in a chessboard pattern. This can utilise almost 100% of the available surface area in the monolith. The smaller the channels, the more larger specific surface area per volume unit.

Small channel opening width however, will also make it more complicated to feed the fluids out/in through the manifold head to or from the monolith's channels. A hole plate system as described above will simplify the feeding into and out of the small channels and will allow fluid distribution in a check pattern to be retained.

In the following, a system is described for feeding two different fluids into and out of monolithic structures without use of a manifold head. The method is based on the fluid channels with the same fluid being arranged in rows in which they share joint walls. In a similar manner to that described in US 4271110, these joint walls can be cut away at a certain depth of the monolith and subsequently be sealed at the end so that openings are created in the sidewalls of the monolith where one of the fluids can be fed in or out.

However, unlike the method described in US Patent 4271110, this method is based on the fluid channels in rows not only running in parallel along the side walls in one direction but a row pattern being formed in both directions (perpendicular to each other). This means that the cuts are made for these intersecting rows, and, after sealing (as described above), the result will be openings in all four side walls of the monolith and not just in two side walls, which is the case when the rows only run in parallel in one direction. This produces greater flexibility for feeding the fluids in and out of the monolith. It will then be possible to arrange the fluid channels in repeating units of 3 x 3 with one fluid in the corner channels and the other fluid in the two centrally intersecting rows (cross). Similarly, it will be possible to have a repeating unit of 4 x 4 channels in which the centrally intersecting connected rows form a cross. The six other channels are then also placed with one in each corner (the top of the cross) and two in the corresponding outer edges on each side at the bottom of the cross.

The present invention makes it possible, in a simple and efficient manner, to feed and distribute two different fluids out of and into individual channels in a multi-channel monolithic structure. This is done by means of a manifold head

that is sealed to the short face or the faces of the monolith where the channel openings are. The method is based on utilising the system in the monolith where channel openings that feed the same fluid are in rows when the two fluids are evenly distributed. The rows of channel openings with the same fluid
5 lead to plenum gaps in the manifold head. The plenum gaps may also be arranged with openings so that the two different fluids can be fed out on either side of the manifold head. This means that we can have separate fluid flows out of or into the individual channels in the monolith from separate plenum gap (i.e. the space formed between two dividing plates). This means that it is not
10 necessary to use pipes to feed the two fluids into or out of the monolith or to make cuts or gaps in the monolith itself. Moreover, it will be possible to stack several monoliths in parallel, i.e. side surface against side surface, and thus feed the fluids out of and/or into an external container through channels formed by inclined walls on the manifold heads. The plenum gaps may also be
15 arranged with slots so that the one of the fluids can be fed in or out on top or on one or both sides of the manifold head while the other fluid is fed in or out from plenum gaps through slots to a tubular space in the manifold head. This means that we can have separate fluid flows out of or in to the individual channels in the monolith from separate plenum gaps (i.e. the space formed between two
20 dividing plates) where the plenum gaps for one of the fluids lead into a tubular space connected to a pipe or circular duct connection.

Moreover, the present invention will make it possible, in the same way as described above, with the stated manifold heads, to distribute two fluids in fluid
25 channels in a chessboard pattern into and/or out of a multi-channel monolith, i.e. with one fluid in the "black" channels and the other fluid in the "white" channels.

If the manifold head is connected directly to the monolith, the distance between
30 the dividing plates in the monolith head will have to be smaller than the channel openings in the monolith. The lower limit of the distance between the dividing plates will therefore determine how small the channel openings may be that are made in the monolith. A system of hole plates between the monolith and the

manifold head will make it possible to feed the fluids into and out of the channels in the monolith that have a size that is much smaller than the distance between the manifold head's dividing plates. In addition, this hole plate system will also make it possible to arrange the fluid channels, which are distributed in a check pattern, in a pattern in which the outlet channels for the same fluid are in one row.

Moreover, a hole plate system between the monolith and the manifold head will make it possible to have a greater distance between the dividing plates than the channel openings in the monolith.

A distribution of the fluid channel openings in a chessboard pattern enables a maximum utilisation of the contact area between the two fluids in the monolith. A plate that covers all the channel openings is sealed to a face of the monolith and to the manifold head. The plate also has a hole pattern equivalent to the channel pattern in the monolith. The channel pattern in the monolith and the hole pattern in the plate are adapted so that holes for the same fluid can form rows of holes over which the plenum gaps are placed.

The present invention requires no processing of the monolith itself if the surface roughness at the channel opening faces meets the tolerance deviation requirements for sealing of the hole plate to the monolith's channel opening faces. If this is not the case, the invention will be usable if the monolith's surfaces are processed, for example surface-ground, to the tolerance deviation requirements for sealing of the hole plate to the channel opening faces.

Through the rows of holes of one fluid in the plate, the fluid is fed in or out through plenum gaps in that which now constitutes a manifold head and out or in through slots in the same manifold head. Accordingly, the other fluid is fed in or out through slots on the opposite side wall of the manifold head or through a tubular connection. The two fluids are thus fed out of their respective channels

in the monolith in such a way that the two fluids can be relatively easily kept apart.

The present invention is explained and illustrated in further detail by means of
5 Figures 1-18.

Figure 1

Figure 1 shows two multi-channel monoliths, both with square cells or channel openings. The monolith on the left hand side has channel walls oriented in parallel with the monolith walls. The monolith on the right hand side has channel walls oriented in 45 degrees angle to the monolith outer walls. Such monolith structures, if made of ceramic materials, will normally be made by means of extrusion. The figure presents the monoliths in perspective from one face showing the channel openings with an exploded view showing the channel details. The extrusion tool determines the monolith's channel structure, cross-section area and shape. A number of different geometric shapes of channels can be made. For example, all the channels cross-section can be triangles, squares or hexagons or there can be combination between these. The channels in a monolith will normally be parallel and uniform in shape along the entire longitudinal direction of the monolith. Monoliths with square channel openings where the channel opening walls are parallel to the sidewalls of the monolith are most common. Monoliths with channel opening walls that are oriented in 45 degrees angle to the outer walls are more unusual. In the present invention such an orientation is preferable because it simplifies the hole pattern and
15 reduce the needed number of hole plates compared to the monolith with channel opening walls parallel with the outer monolith wall.

Figure 2

Figure 2 displays an assembly of a monolith with hole plates and manifold head. Typical a monolith stack or monolith unit will have two such manifold heads at the two monolith faces where we find the inlet and outlet openings of the channels. By means of the hole plates fluid flow system is transformed from
30

linear arrangement in manifold head to chessboard pattern arrangement in monolith or vice versa. The manifold head is built up by a set of dividing plates (partition plate A and partition plate B) and two end covers; type "A" and type "B". As can be seen from figure fluid 1 can enter or leave through tubular openings inside the manifold head. In Figure 2 the tubular openings is in the centre position of the manifold head, but in principle any position within manifold head can be used. Also the shape of the manifold head is flexible besides the face that fits to the converter plates or directly to the monolith faces where we find the inlet and outlet openings of the channels. The tubular opening makes it possible to connect to a neighbouring monolith stack with a similar manifold head through a tubular connection or connect manifold head to a collecting pipe for a number of monolith stacks. Thus fluid 1 can be fed in and out through a closed piping system to and from a number of monoliths while the other fluid enter or leave through opening slots in the manifold head. Such a solution is advantageous for a system where the monolith stacks are placed inside a pressure vessel because only one of the fluids (here fluid 1) needs to be hermetically sealed where the other fluid (here fluid 2) is allowed to fill empty space in pressure vessel and flow through ducts or channels to or from inlet and outlet openings in the vessel shell.

The first hole plate sealed to the monolith faces where we find the inlet and outlet openings of the channel have openings (holes) that correspond to the number of channel openings in the monolith. The holes are arranged with openings that are positioned above the monolith channel opening such that two fluids can flow from monolith channels to the gap between the dividing plates in the manifold head and vice versa. For the functionality of the system the openings for one fluid in the plate sealed to the monolith (arranged in chess pattern for maximum area utilisation) must be led through a set of connected openings in a set of connected plates that change position of the fluid flow in such a way that the same fluid is led out through a linear pattern of openings that fits within the openings between the partition plates that is for the same fluid.

Figure 3

Figure 3 shows the front view of one monolith with the channel openings together with five hole plates. Plate 1 has holes with a pattern that is made in such a way that each hole has a position that correspond with the position of one channel opening in the monolith. When plate 1 thus is placed above the monolith in correct position each hole should correspondingly fit within a monolith channel opening. Plate 1 can be sealed to the monolith in this position. The diameter of the holes in plate 1 is most preferable somewhat smaller than the width of the channel openings. How much smaller depends on tolerances and pressure drop that can be accepted. By tolerances here is meant the deviation in shape and size that can arise during production. For ceramic materials one of the reasons for deviations is the shrinkage that arise during sintering of the material. Smaller holes give larger tolerances and larger deviations can be accepted. On the other hand smaller openings in plate 1 will give larger pressure drop for a fluid flowing through. Plate 2,3 and 4 here named the middle plates have holes with longitudinal shapes. This shapes ensures that the fluids can change position from a chess flow arrangement in the monolith to a linear flow arrangement when led out through the holes in plate 5. The stapled lines show the position of the dividing plates of the manifold head. The fluid converter system managed through holes in the plates can also be made with fewer plates or even with one plate. If made up with one plate one need a production technique that enables making small channels leading the outlet or inlet fluid to the correct position. That is either the openings corresponding to the monolith or to the openings corresponding to the position between the partition plates. Injection moulding can be such a method, but very strong demand is put on the technique due to the small tolerances given by the very narrow channels with small distances between each other. One believe that at least making plate 1 and 5 as individual plates gives better control as they can be sealed directly to monolith and partition plates.

Figures 4.1 and 4.2

Figure 4.1 shows a section view of the manifold head with arrows indicating fluid flow direction. The fluids are fed into or out from the monoliths through slots that allow fluid 1 to enter from the circular opening ("tunnel") to the enclosed space (gap) between the dividing plates that separate fluid 1 from fluid 2. As displayed the dividing plates for fluid 2 have no opening into the circular space, but opening slots on top of the manifold head and thus fluid 2 can enter through these slots. Thus fluid 1 and fluid 2 can come from and let into separated plenum chambers or gaps between the dividing plates. The openings from the circular space for fluid 1 are made because the dividing or partition plate B have a set of boss near the circular opening. They will increase the ability of the dividing plates to withstand pressure differences and they can also transfer axial force required for a sealing ring if two or more manifold heads are to be coupled together.

Figure 4.2 shows a manifold head of same system as Figure 4.1, but with two tubular openings inside manifold head. With such a system both fluids can be fed in and out from the monolith in a hermetically closed or sealed piping system. One then can keep the monolith structures in an insulated vessel at atmospheric conditions even if both fluids are at elevated pressures. The disadvantage is that movement due to thermal expansions are restricted by the tubular connections of both fluids.

Figure 5

Figures 1 - 4 deal with an individual system of one monolith with its manifold head.

Figure 5 shows a system for coupling two or more monolith stacks. By means of a sealing ring, an end cover type "A" from one manifold head and an end cover type "B" from another manifold head and an axial force two monolith stacks can be coupled together (see Figure 6). Such a system is special applicable in industrial processes where often a large number of monoliths is needed.

Figure 6

Figure 6 illustrates the coupling principle between two manifold heads, showing sealing ring and two types of end covers; type "A" and "B". The contact surface between the sealing ring and end cover "A" is a plane surface that permits 2-axis movement on the surface. The contact surface between the sealing ring and end cover "B" is a part of a spherical surface that permits rotation around the centre of the sphere. Note the external force that is applied to the manifold head. This force is necessary to make the system gastight, especially if "Fluid 1" has higher pressure than "Fluid 2". If "Fluid 2" has sufficient overpressure compared to "Fluid 2", an external force should not be necessary.

The circular exploded view in Figure 6 shows the sealing ring and the two different types (type A and B) of end covers used to connect manifold head of one monolith stack with manifold head of another neighbouring monolith stack. With such a system one can do the coupling of two different monoliths in such a way that both fluid tightness and flexibility in movement can be maintained. Another aspect is that with such a system the coupling of the two monolith stacks can be done in a very compact way. The only distance is the required thickness of the sealing ring.

Figure 7

Figure 7 is explaining the spherical contact surface between sealing ring and end cover "B". This figure shows how the contact surface between the sealing ring and end cover "B" is a part of a spherical surface that permits rotation around the centre of the sphere.

Figure 8

Figure 8 shows an assembly with two monoliths and manifold system connected to each other. The expanded view shows placement and details of the couplings described in Figures 5-7.

Figure 9

Figure 9 shows an alternative converter design using a monolith with a cell pattern oriented in 45 degrees compared to the monolith wall. Such a monolith needs maximum four hole plates compared to the solution in Figure 3 that need five. Also the space or distance between dividing plates is increased compared to the method or system shown in Figure 3, given the same monolith cell size. The lower right part of Figure 9 shows the cavity. The cavity is what is left when all the material is taken away. The cavity of the "flow channels" inside the four hole plates can be seen.

Figure 10

Figure 10 shows an individual monolith stack consisting of the monolith, the converter plates and manifold heads. Connector plates are also shown. Such plates is included only if the monolith stack is made up of two or more individual monoliths. This can be the case if the length of one individual monolith is not sufficient or because the system consists of monoliths with different functionalities or properties. E.g. one monolith can be a heat exchanger and the other monolith can consist of a membrane structure. The connectors can consist of a graded material such in case of different thermal expansions of the monoliths both can be matched.

Figure 11

Figure 11 shows a row of monolith stacks consisting of individual monolith stacks coupled together. To build up such a line of monolith stacks the coupling system shown in Figure 8 can be used. If scaling up to industrial sizes one will start with the smallest repeating unit which for this system would be the individual monolith stack as shown in Figure 10. The next unit component is an assembly or a line of monolith stacks coupled together as shown in Figure 11.

Figure 12

In large industrial scale applications, where many hundreds monoliths have to be used, it is of importance that the monolith stacks can be arranged close

together for compact reactor design solutions. Figure 12 shows a system or method where line of monolith stacks as shown in Figure 11 are stacked wall to wall constituting one large "monolith block". In Figure 12 one line or row consists of ten monolith stacks. How many stacks there should be pr row depends on several factors. To fit into a cylindrical pressure vessel for maximum utilisation of volume the height of the stack and width of the monolith block should correspond. Thus a stack height of 150 cm the row should consist of 10 monoliths if width of manifold head and monolith is 15 cm. The capacity of the system can then be increased without increasing diameter of pressure vessel by simply increasing length and adding number of monolith stacks.

Figure 13

Figure 13 shows the arrangement of the monolith block within a cylindrical pressure vessel. As can be seen the number of rows can be increased or decreased without changing the diameter of the pressure vessel. Thus system can simply be adjusted to a wide range of capacities by changing the number of rows and adjusting length of pressure vessel. In Figure 13 fluid 1 is kept in a closed system by means of internal inlet and outlet collecting pipes. In Figure 13 a counter-current flow system in the monoliths is shown were fluid 1 entering top manifold head in the monolith stacks flow downwards and is led out in the bottom manifold head. Fluid 2 enter lower manifold head from ducts or open space inside reactor vessel and flow upward in the monolith channels and out in the upper manifold head and into the top of the reactor were it is led out through the opening slots in the manifold heads in the upper part of the reactor.

Figure 14

Figure 14 shows the monolithic structures inside a pressure or reactor vessel. In this system fluid 2 is fed in and out at the same position on the pressure vessel wall. This system could e.g. be adapted when fluid 2 comes from a compressor and fluid 2' is led out to a turbine. Fluid 2 could be air and fluid 2' could be oxygen depleted heated air. The monoliths can be ceramic oxygen delivering membranes and fluid 1 is the permeate fluid that receives the oxygen from air. Fuel could then be injected in fluid 1 and a combustion will take place

consuming the oxygen and heat will be produced. With such a system the oxygen depleted fluid 1 (after combustion) could be returned to the monoliths with walls consisting of an oxygen transferring membrane. Fluid 1 is heated by combustion and heat is transferred from fluid 1 to the oxygen containing fluid 2.

- 5 At a defined temperature level the membrane in the monolith wall transfer oxygen to fluid 1. The surplus mass due to injected fuel and oxygen can be led out as bleed gas through the monolith on the left side to a collector pipe. The monolith on the left side can then be used as a pure heat exchanger; heating air and cooling bleed gas. If fluid 1 consists of water vapour and carbon dioxide
- 10 such a design or system solution can be used for gas power production with CO₂ handling. A zero emission power plant can then be made if CO₂ is sent to permanent storage.

Figure 15

- 15 Figure 15 is a cross sectional view of the reactor shown in Figure 14. This figure illustrates the process flow system by using arrows showing flow direction. One can see how inlet fluid 2 is led by ducts close to the inner wall and in to the lower part of the reactor where it enters the lower manifold heads of the monolith stacks. Fluid 1 flows counter current to Fluid 2 in a circulation loop. For
- 20 the system of zero emission gas power fluid 2 is air and monoliths are ceramic oxygen membranes. Fluid 1 component can be water vapour and carbon dioxide, which then receives oxygen from air. A fuel like natural gas is then added for combustion and fluid 1 can then be returned to monoliths to receive oxygen (flux is driven by oxygen partial pressure difference) and heat Fluid 2
- 25 and 2' leaving for the power generating turbine. To ensure a mass balance in the fluid 1 circulation loop a bleed is taken out. Thus the left monolith stack has a functionality of a pure heat exchanger. Fuel injection can be done by mean of a fuel ejector to ensure circulation of fluid 1.

Figure 16

- 30 Figure 16 shows a reactor concept for combined production of oxygen and power where the monoliths are made of oxygen transporting membranes. This

illustrates the flexibility of the present invention for utilisation of different process systems.

With only minor modification the same reactor concept as shown in Figures 14 and 15 can be used to combine oxygen and power production. Fluid 2 can be compressed air, which is heated in the bottom of reactor by means of gas burners. Thus some of the oxygen in air is consumed to heat the air to a temperature suitable for the ceramic oxygen transporting membranes. Fluid 1 must have a lower partial pressure of oxygen than in fluid 2. The lower partial pressure ensures that oxygen is transported from fluid 2 to fluid 1 through the membrane. It is also possible to use vacuum to pull out the oxygen on the permeate side of the membrane instead of a fluid 1. This will directly produce pure oxygen that can be compressed to delivery or storage pressure.

For maximum power generating capacity oxygen left in fluid 2 at the outlet of membranes can be used for increasing the temperature of air to turbine by having gas burners in outlet duct or pipe as shown on figure. Fluid 1 can in principle be any fluid (and even air at lower pressures than in fluid 2 ensuring oxygen positive partial pressure difference) capable of transporting oxygen out from the membrane and suitable for downstream separation from oxygen or direct applications.

Figure 17

Figure 17 shows the system assembly of monolith, hole plates and manifold head. In the illustrated manifold head the outlet opening (here for fluid 2) have a shorter distance and a straighter direction than the manifold head in figure 2. The dividing plates have guiding ribs for fluid 2 also performing mechanical support. The ribs are shaped to prevent blockage of the holes and minimise flow restriction for fluid 2. Fluid 1 has a circular inlet to the manifold head and open slots where fluid 1 can enter through the hole plates and in to the monolith channels. There is no ribs or boss on the fluid 1 side of the dividing plate. In Figure 9 a system of four individual plates for transferring the fluids are shown

compared to only 2 in figure 17. The plates of figure 17 holds the same functionality as the four plates of Figure 9. Plate 1 corresponds to plate 1 of Figure 9 and plate 2 corresponds to plates 2-4 of Figure 9.

5 **Figure 18**

Figure 18 shows detail views inside plate 2 and plate 1. The thickness of plate 2 is dependant on the sloping angle of the funnel leading to the opening holes in plate 1 for fluid 1 and fluid 2 as well as the number of holes from plate 1 each funnel shall collect. As can be seen from the exploded view on the left hand
10 side the funnel for fluid 2 collects from four holes from plate 1 and thus also from four channels from the monolith. The exploded view on the right hand side shows the funnel for fluid 1 and as can be seen these funnels collect or distribute to five holes in plate 1. Due to symmetry one have made an even number of holes for each funnel. Every fifth hole has then to be distributed to
15 two funnels. Figure 18 illustrates only a principle design of plate 2. Thus all kinds of combinations between the number of holes that each funnel shall collect from or distribute to can be chosen freely. The selected combination will depend on a set of parameters among them pressure drop, the number and distance between dividing plates.

20

The present invention offers possibilities for improvement and simplification of unit operations for heat and mass transfer (separation) by utilising the monolithic structures' compactness (i.e. large surface area per volume unit with small channels), low flow resistance for gases and high-temperature resistant
25 ceramic material, which can be coated with a catalyst. The improvements will be associated with use of the monoliths in mass and heat transfer between two different fluids and the fact that these unit operations in the monolithic structure can be integrated with a chemical reaction. Such a combination of mass and heat transfer and chemical reaction (unit operations) in the monoliths will
30 contribute to producing compact solutions in which transport and separation are simplified. One application will be a combination of endothermic and exothermic reactions, for example steam methane reformation of natural gas or other

substances containing hydrocarbons to syntheses gas (hydrogen and carbon monoxide) with endothermic steam methane reformation in catalyst-coated channels and exothermic combustion in adjacent channels. Such monolithic structures can produce very compact reformers and can, for example, be used for small-scale hydrogen production. However, syntheses gas can also be processed further into a number of other products, for example methanol, ammonia and synthetic petrol/diesel.

Higher operating temperatures where metals not can be used (800-900°C and above) are favourable in terms of equilibrium or thermodynamics by many chemical processes. In such processes, ceramic monoliths, which can both be coated with catalyst and tolerate higher temperatures, can be very advantageous. Thus a combustion or hot gas process can directly be combined with a chemical reaction process.

Monolithic structures can also be used in the energy market (power production), for example for catalytic combustion of natural gas. By utilising the present invention the temperature window of the combustion process can be controlled resulting in reduced nitrogen oxide (NO_x) production. Combustion or oxidation in air or any atmosphere where oxygen and nitrogen is present always will have the potential of producing NO_x. This environmental harmful gas is mainly produced in the high temperature zones of the combustion flame. By utilising the present invention with chessboard flow gas distribution in the monolith, one can have catalytic combustion of a mixture of fuel and air producing heat in the "black" channels and a passive coolant (i.e. air) in the "white" channels or an active coolant performing an endothermic reaction (i.e. steam methane reforming) in the "white" channels. Such a system will prevent peak temperatures and thus reduce NO_x production. Furthermore, with this system one have the possibility to mix coolant and combustion gas downstream monolith by having a manifold only in inlet position (given co current flow) and thus a very efficient mixing in outlet position due to chessboard pattern and small channels in the monolith.

The system described above for preventing NO_x formation can also be used for preventing/reducing emission of other unwanted components. Thus the present invention can combine combustion (heat production) and heat transfer directly in monolith structures through the thin contact wall between two fluids.

Claims:

1. A method for distributing two fluids into and out of the channels in a multi-channel monolithic structure where the channel openings are spread over the entire cross-sectional area of said structure and said channels have joint walls,

characterised in that

one fluid is fed through a slot in one or more gaps in a manifold head which is sealed to one face of said monolith structure,

the other fluid is fed into a tunnel in said manifold head and further through slots in said tunnel wall and into one or more gaps in said manifold head,

said fluids are distributed from their respective gaps into said channels in such a way that at least one channel wall is in common for said fluids,

said fluids are collected in their respective gaps in a manifold head which is sealed at the opposite side of said structure where the first manifold head is sealed,

the fluids are then respectively led through a slot from one or more gaps and slots in a tunnel wall in said last mentioned manifold head.

2. A method for distributing two fluids into and out of the channels in a multi-channel monolithic structure where the channel openings are spread over the entire cross-sectional area of said structure and said channels have joint walls,

characterised in that

one fluid is fed into a first tunnel in a manifold head and through slots in said first tunnel wall and further into one or more gaps in said manifold head,

the other fluid is fed to a second tunnel in said manifold head and through slots in said second tunnel wall and further into one or more other gaps in said manifold head,

said fluids are distributed from their respective gaps into said channels in such a way that at least one channel wall is in common for said fluids,

said fluids are collected in their respective gaps in said manifold head, the fluids are then led out of their respectively slots in said tunnels walls.

3. A method according to claim 1 or 2,

characterised in that

said fluids are fed into and out of the same manifold head.

4. A method according to claims 1-3,

characterised in that

said fluids are distributed in said channels in such a way that one fluid flowing in a channel has the other fluid flowing in all the adjacent channels.

5. A method according to claims 4,

characterised in that

said fluids from said gaps are distributed in said channels as in a checkboard pattern with one fluid in the "black" channels and the other fluid in the "white" channels.

6. A manifold head for distribution of two fluids into and out of the channels in a multi-channel monolithic structure where the channel openings are spread over the entire cross-sectional area of said structure and where said channels have joint walls,

characterised in that

said manifold head comprises:

at least three parallel dividing plates joined together with spacers to form gaps with slots between said plates and

end cover plates joined in parallel to said dividing plates where said dividing plates and cover plates have one opening forming a tunnel with slots through said joined plates.

- 5 7. A manifold head according to claim 6,
 characterised in that
 said dividing plates and cover plates have at least one hole each
 forming a tubular space (tunnel) through said joined plates and where
 said tunnel wall has slots communicating with said gaps

10

8. A unit,
 characterised in that
 said multi-channel unit comprises:
 a monolithic structure where the channel openings are spread over the
 entire cross-sectional area of said structure and said channels have
15 joint walls and a manifold head according to claim 6 or 7 which is
 sealed to at least one face of said structure.

15

9. A unit,
20 **characterised in that**
 said unit comprises:
 a multi-channel monolithic structure where the channel openings are
 spread over the entire cross-sectional area of said structure and said
 channels have joint walls,
25 a manifold head according to claim 6 or 7 which is sealed to at least
 one face of said structure,
 and at least one hole plate which is sealed between said manifold head
 and said structure on said face where the channel openings are.

25

- 30 10. A unit according to claim 9,
 characterised in that
 said holes are arranged in such a way that two fluids can flow from said
 monolith channels to said gaps and vice versa.

30

11. A unit according to claim 8 or 9,
characterised in that
one or more of said channel walls are coated with one or more catalytic active components.

5

12. A unit according to claim 8 or 9,
characterised in that
said channel openings are evenly distributed over the entire cross-sectional area of said monolith structure as in a chessboard pattern.

10

13. A unit according to claim 8 or 9,
characterised in that
said structure has channel walls oriented in 45 degrees angle to the outer structure walls.

15

14. A unit according to claim 8 or 9,
characterised in that
said dividing plates are sealed to a hole plate.

20

15. A unit according to claim 8 or 9,
characterised in that
said dividing plates are sealed directly to the monolith channel walls.

25

16. A unit according to claim 8 or 9,
characterised in that
said manifold head is sealed to at least one face of the monolith structure where the channel openings are.

30

17. A stack,
characterises in that
said stack comprises:

two or more multi-channel monolithic structures where the channel openings are spread over the entire cross-sectional area of said structures and said channels have joint walls,
at least one manifold head according to claim 6 or 7 which is sealed to
5 at least one face of said structure,
at least one plate with holes which is sealed between said manifold head and said structure on said side where the channel openings are,
and at least one connector plate or other coupling device between units.

10

18. A row of units or stacks,
characterised in that
said row comprises
units according to claims 8-16 or stacks according to claim 17 coupled
15 together.

19. A row of units or stacks,
characterised in that
said row comprises
20 units according to claims 8-16 or stacks according to claim 17 where a
sealing ring and two different types (type A and B) of end covers are
used to connect said manifold head of one unit or stack with said
manifold head of another neighbouring unit or stack.

25 20. A block,
characterised in that
said block comprises rows of units or stacks according to claim 18 or
19 which are stapled face to face.

21. A reactor for mass and/or heat transfer between two fluids,
characterised in that
one or more of the units according to claim 8-16 or stack according to
5 claim 17 or row of units or stacks according to claim 18 or blocks
according to claim 20 is integrated in said reactor.

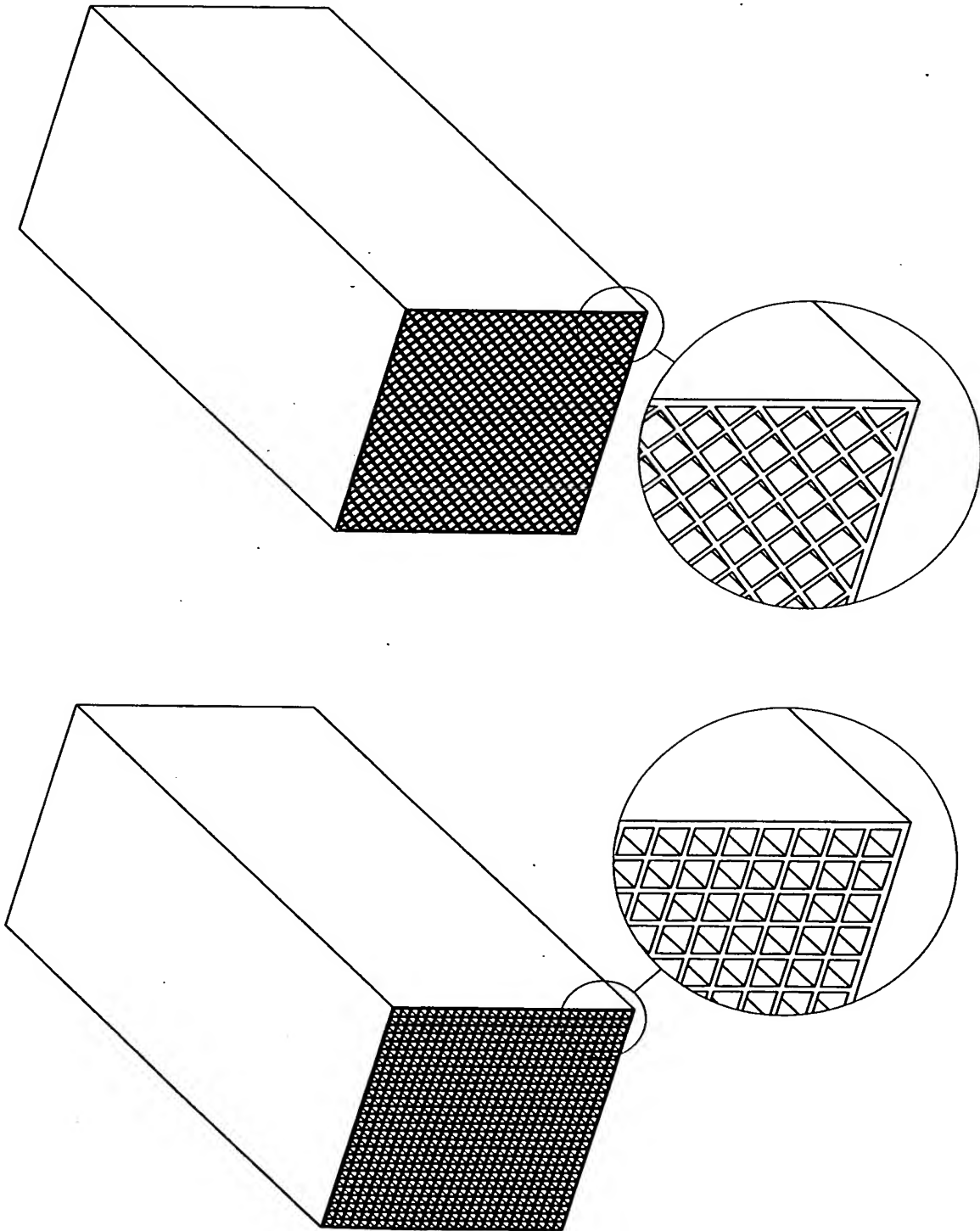
22. A method for mass and / or heat transfer between two fluids,
characterised in that
10 said two fluids are distributed through one or more units according to
claim 8-16 or stack according to claim 17 or row of units or stacks
according to claim 18 or blocks according to claim 20.

Abstract:

5 The present invention concerns a method and equipment for distribution of two fluids into and out of the channels in a multi-channel monolithic structure (monolith) where the channel openings are spread over the entire cross-sectional area of said structure.

Said equipment consists of a manifold head, a monolith unit or a monolith stack, or a row of said units or stacks or a monolith block.

10 Furthermore, the present invention concerns a method and a reactor for mass and/ or heat transfer between two fluids where said fluids are distributed through one or more of said manifold heads and units or stack or row of units or stacks or blocks.

**Fig. 1**

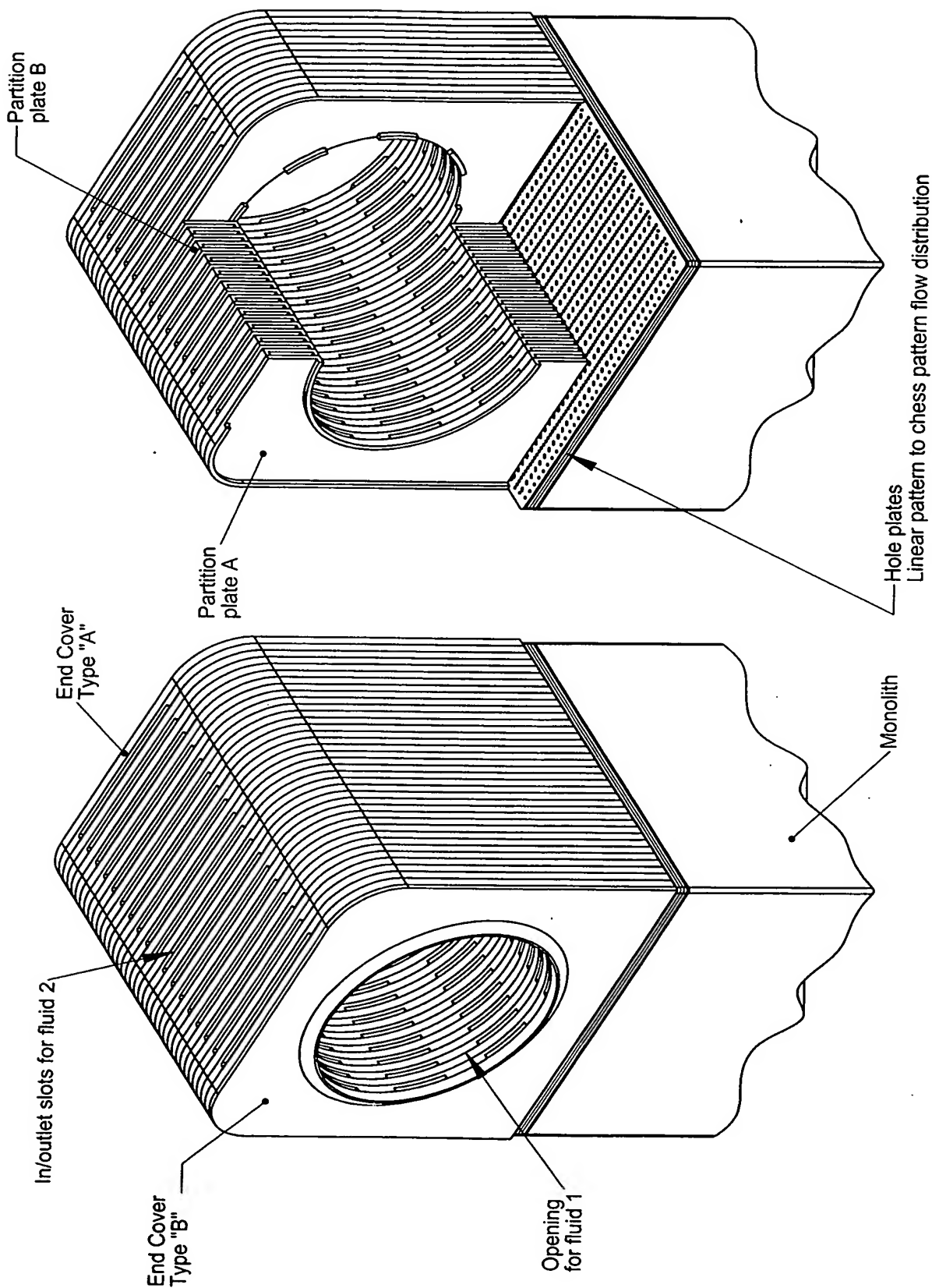


Fig. 2

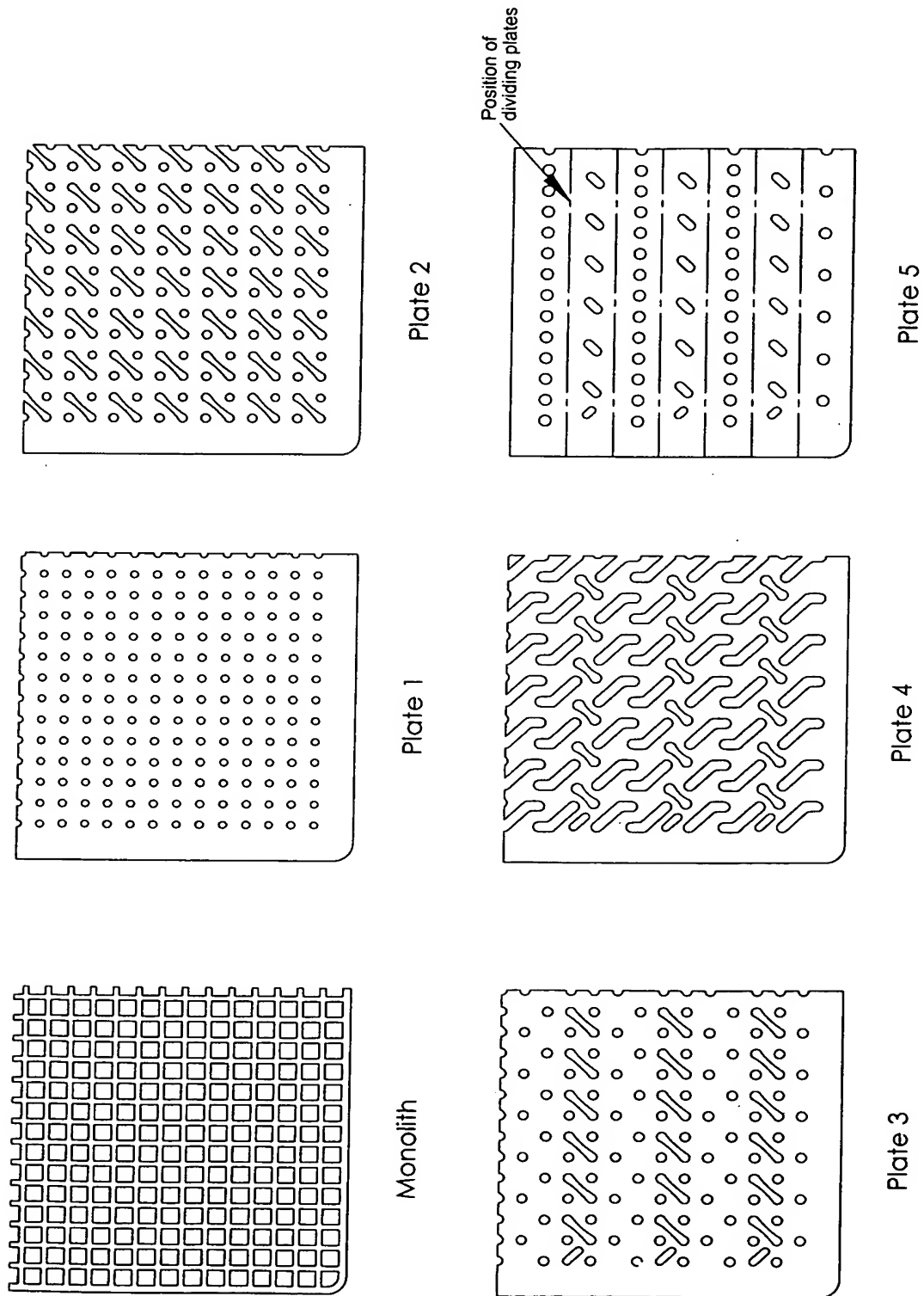


Fig. 3

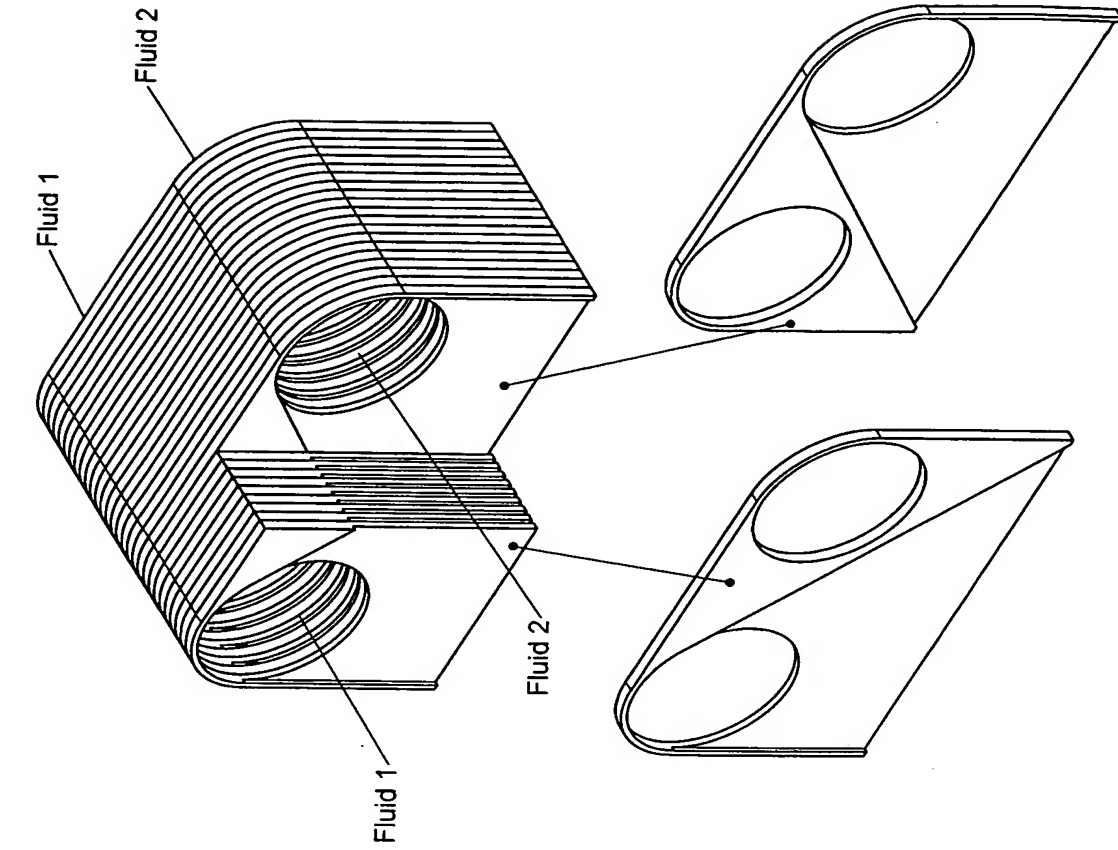


Fig. 4.1

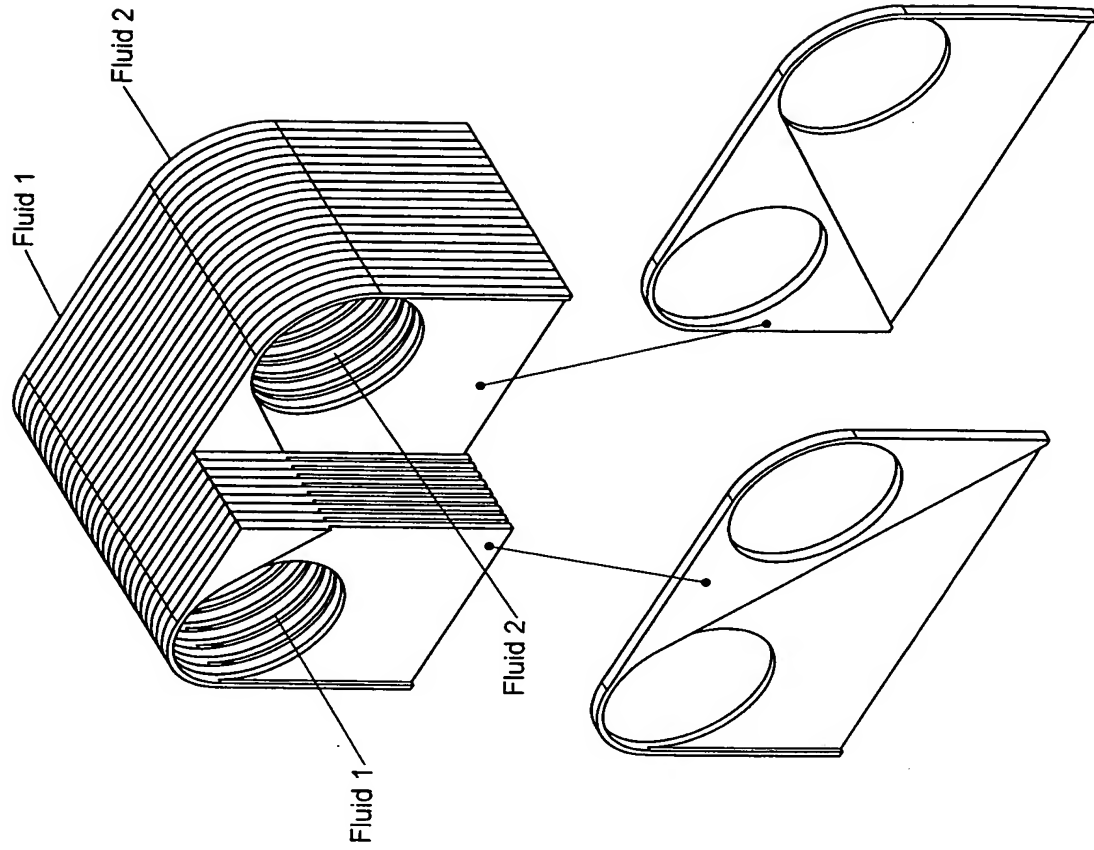


Fig. 4.2

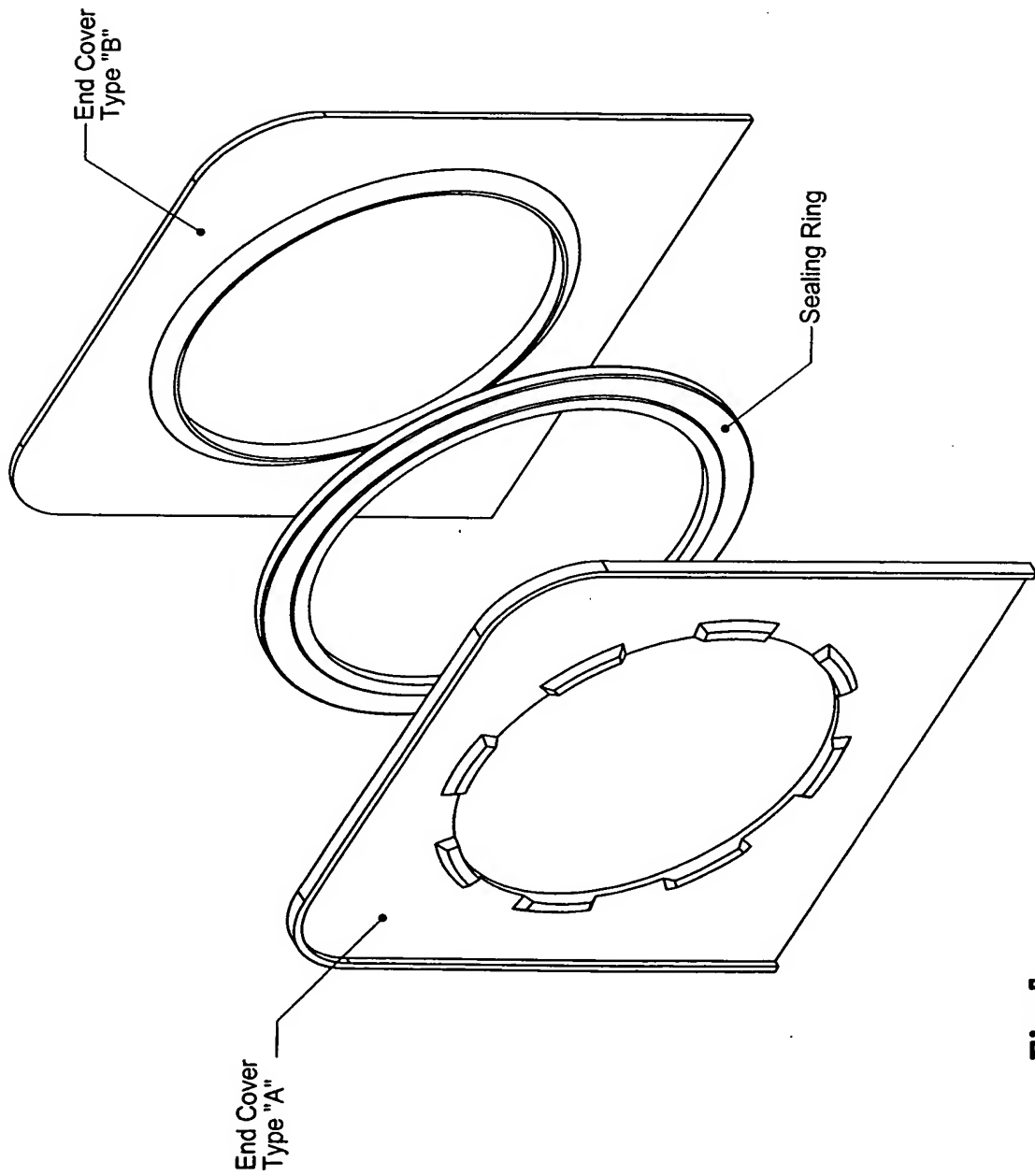


Fig. 5

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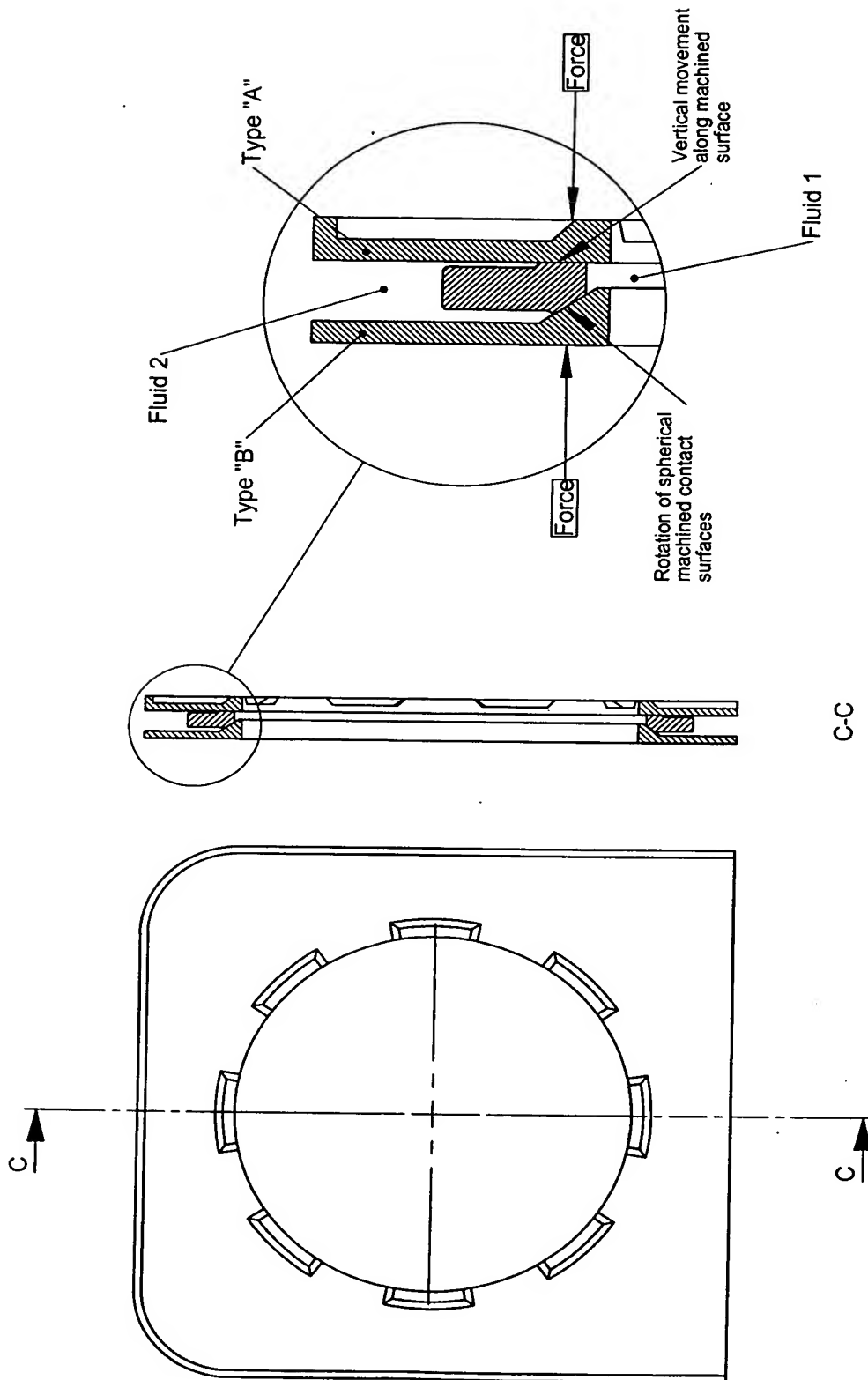


Fig. 6

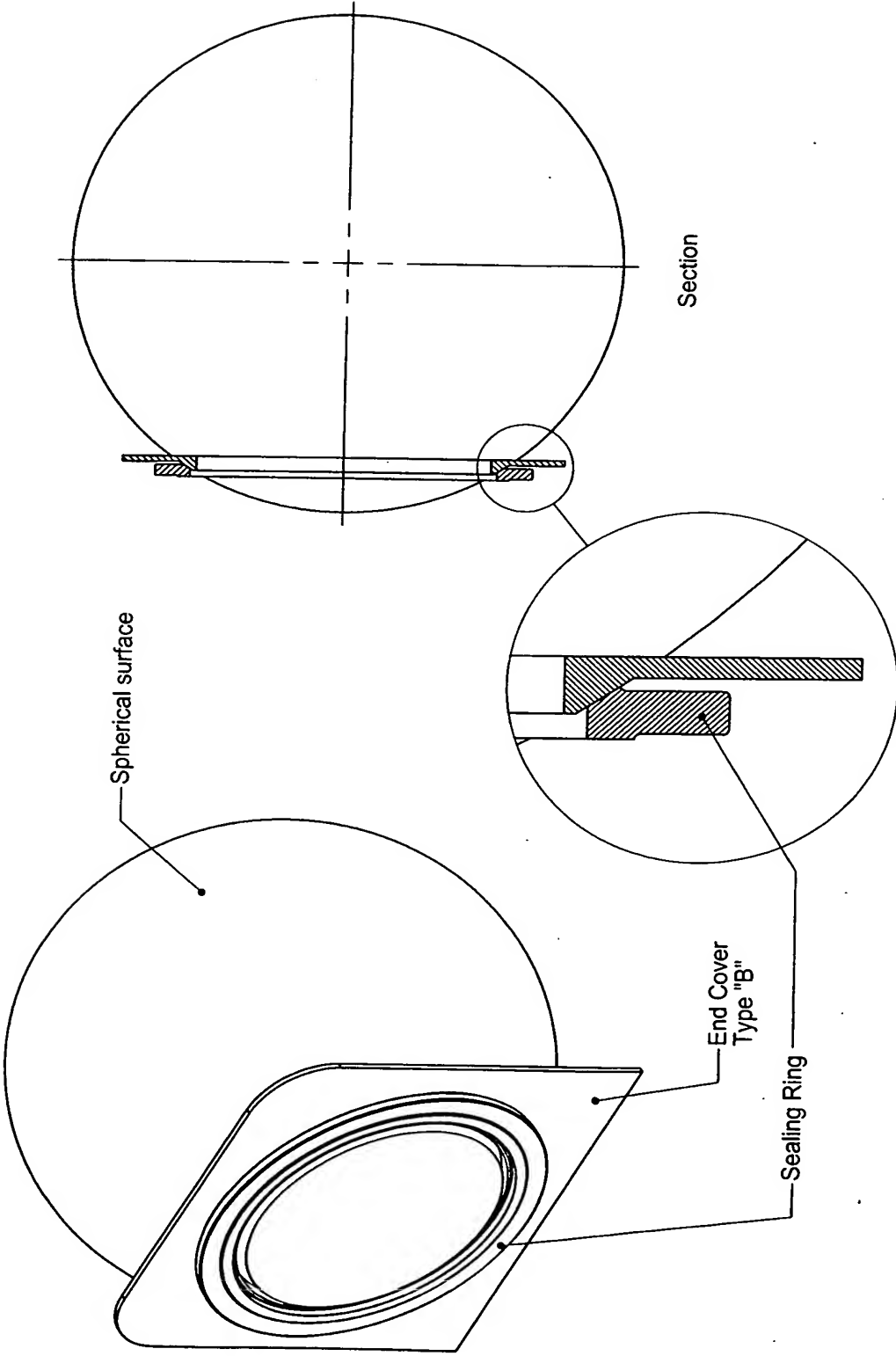


Fig. 7

8/18

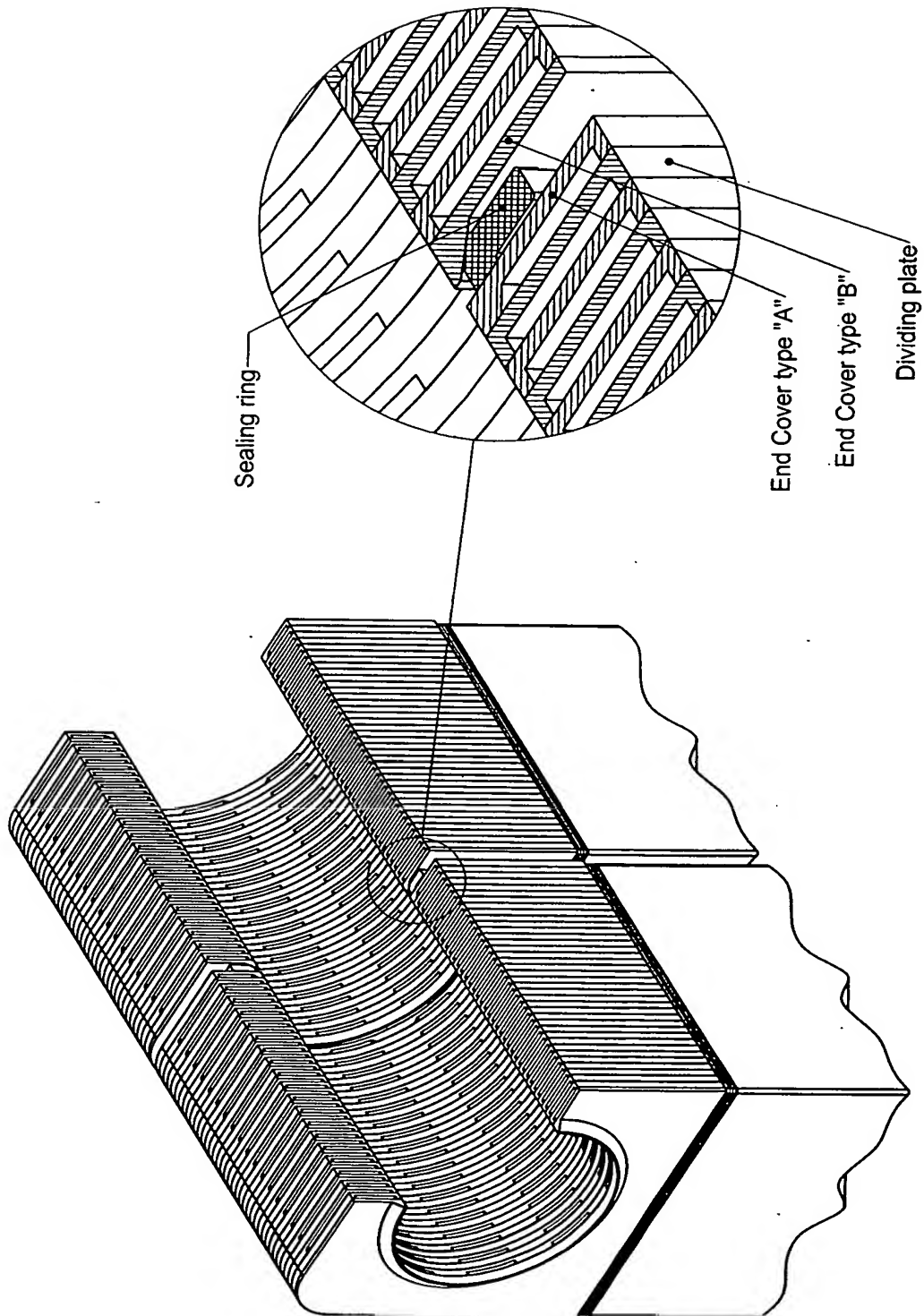


Fig. 8

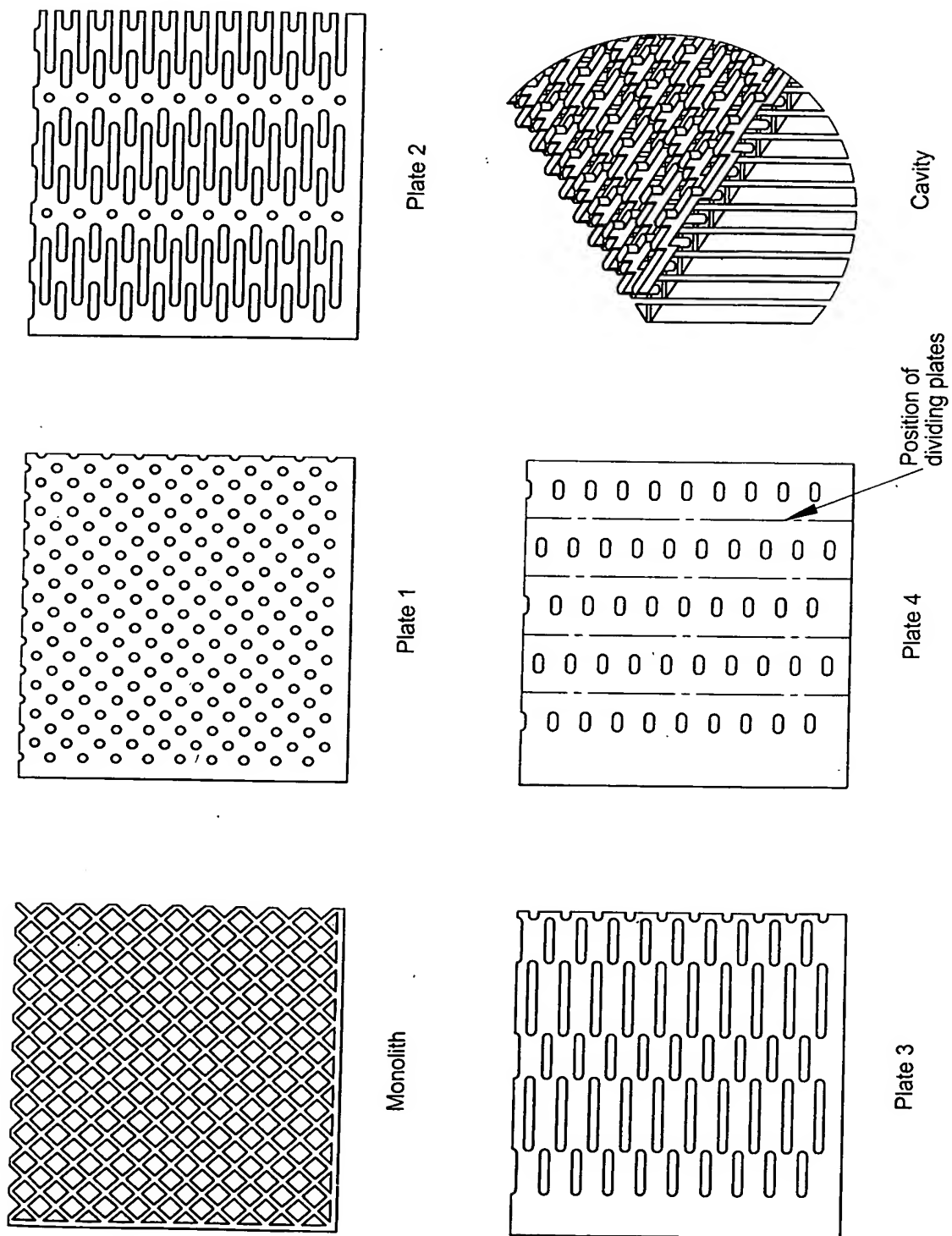
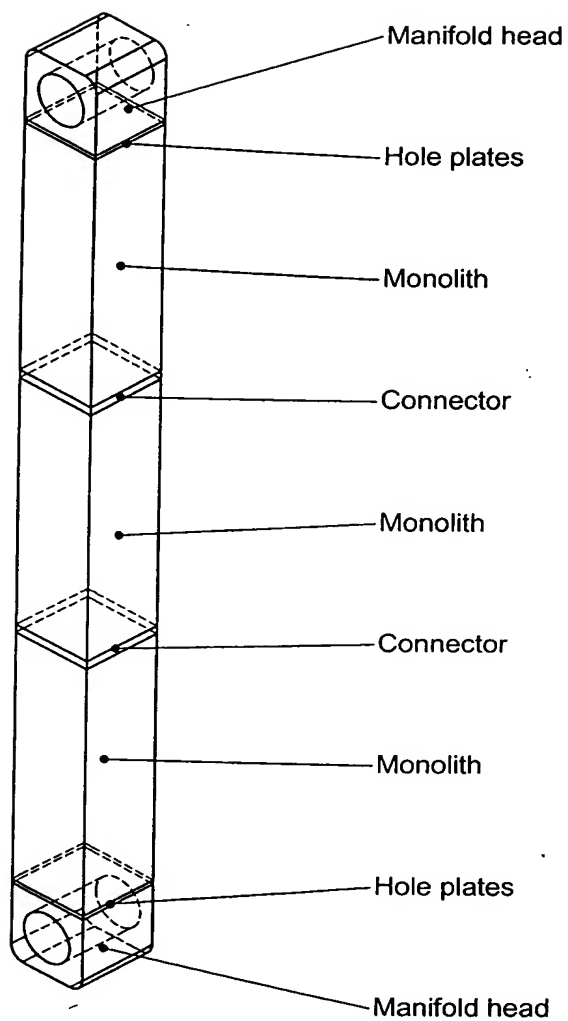


Fig. 9

**Fig. 10**

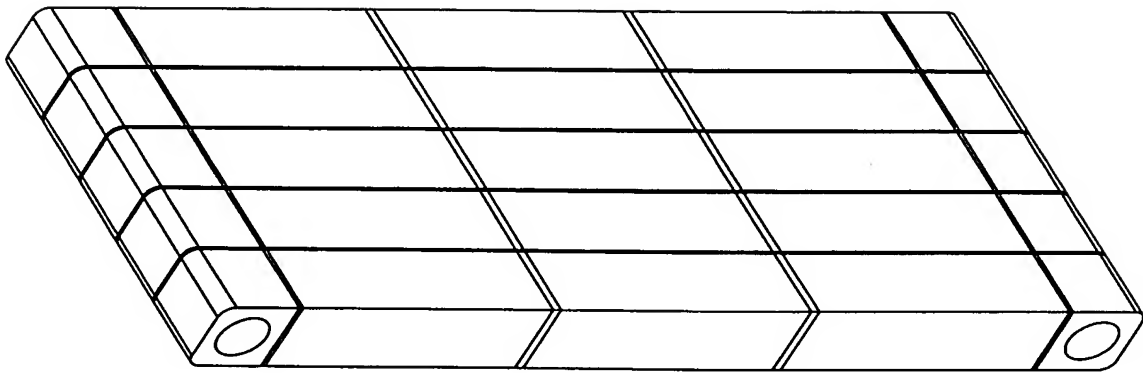


Fig. 11

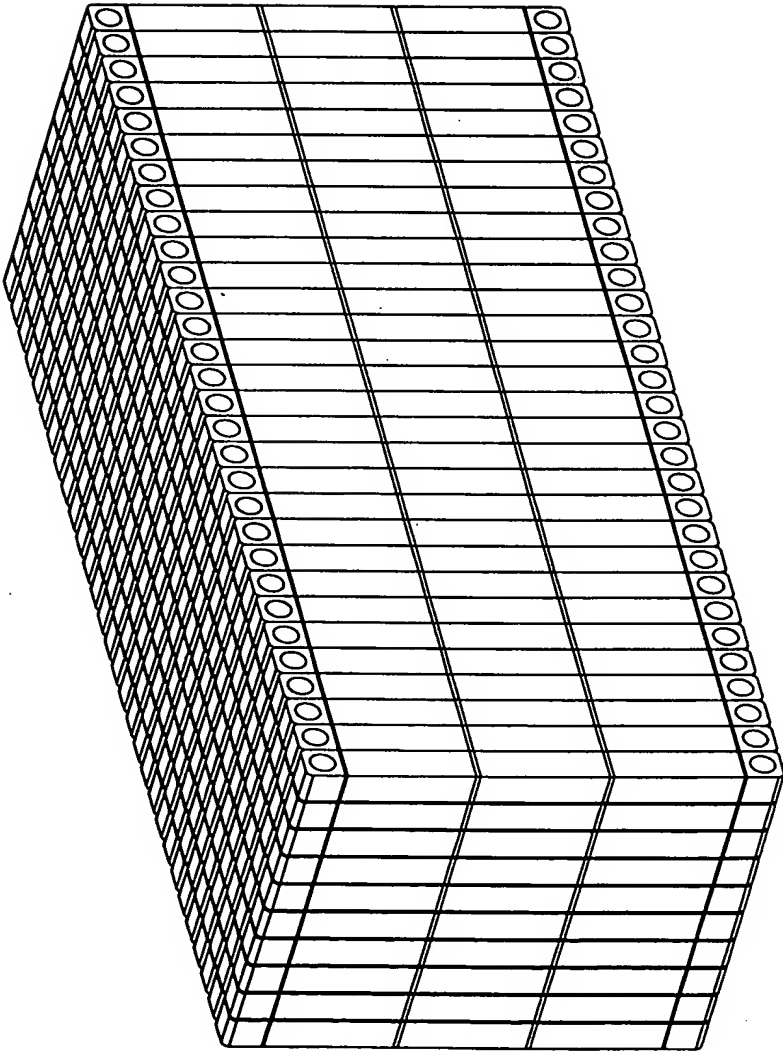


Fig. 12

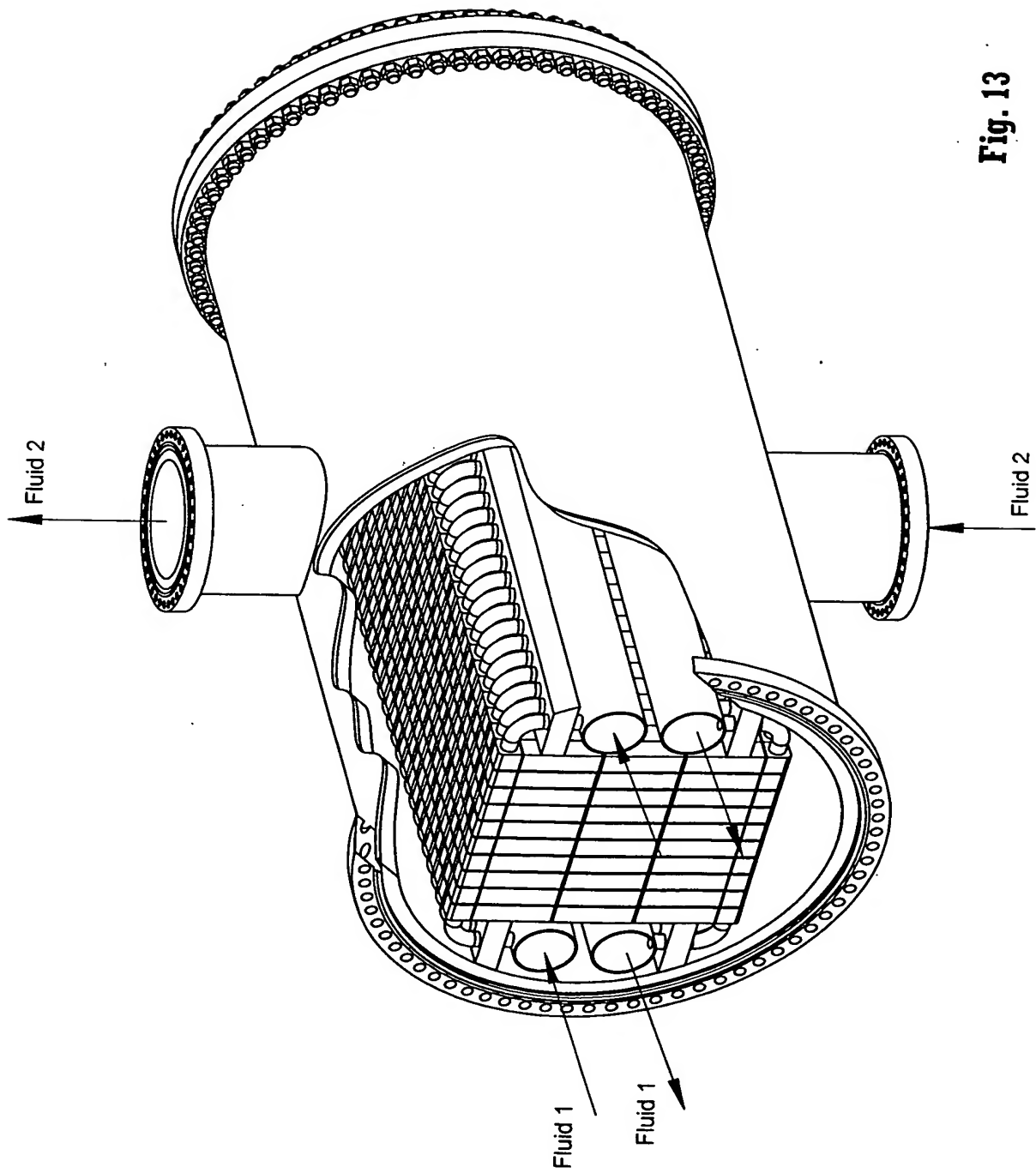
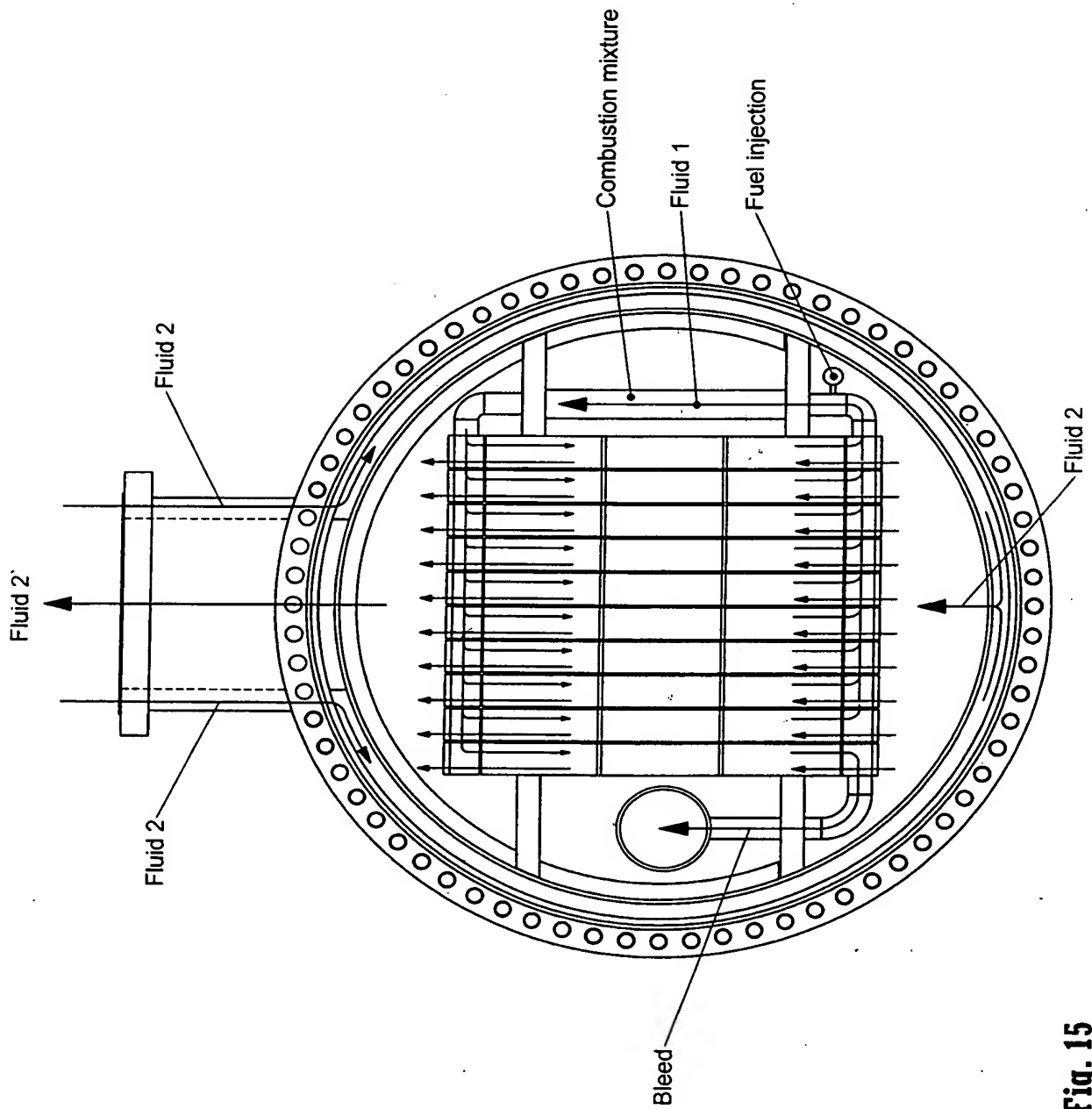




Fig. 14

15/18



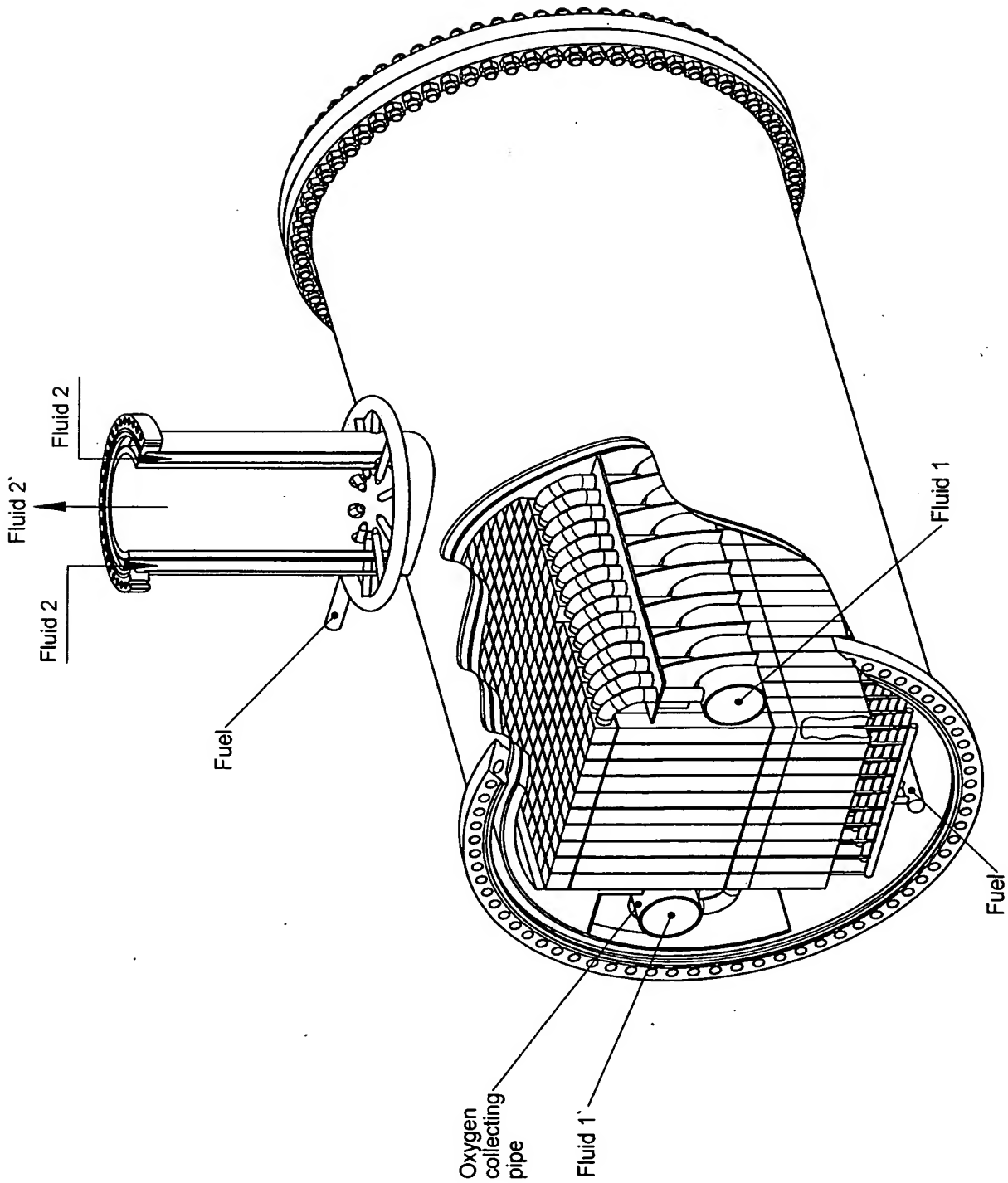
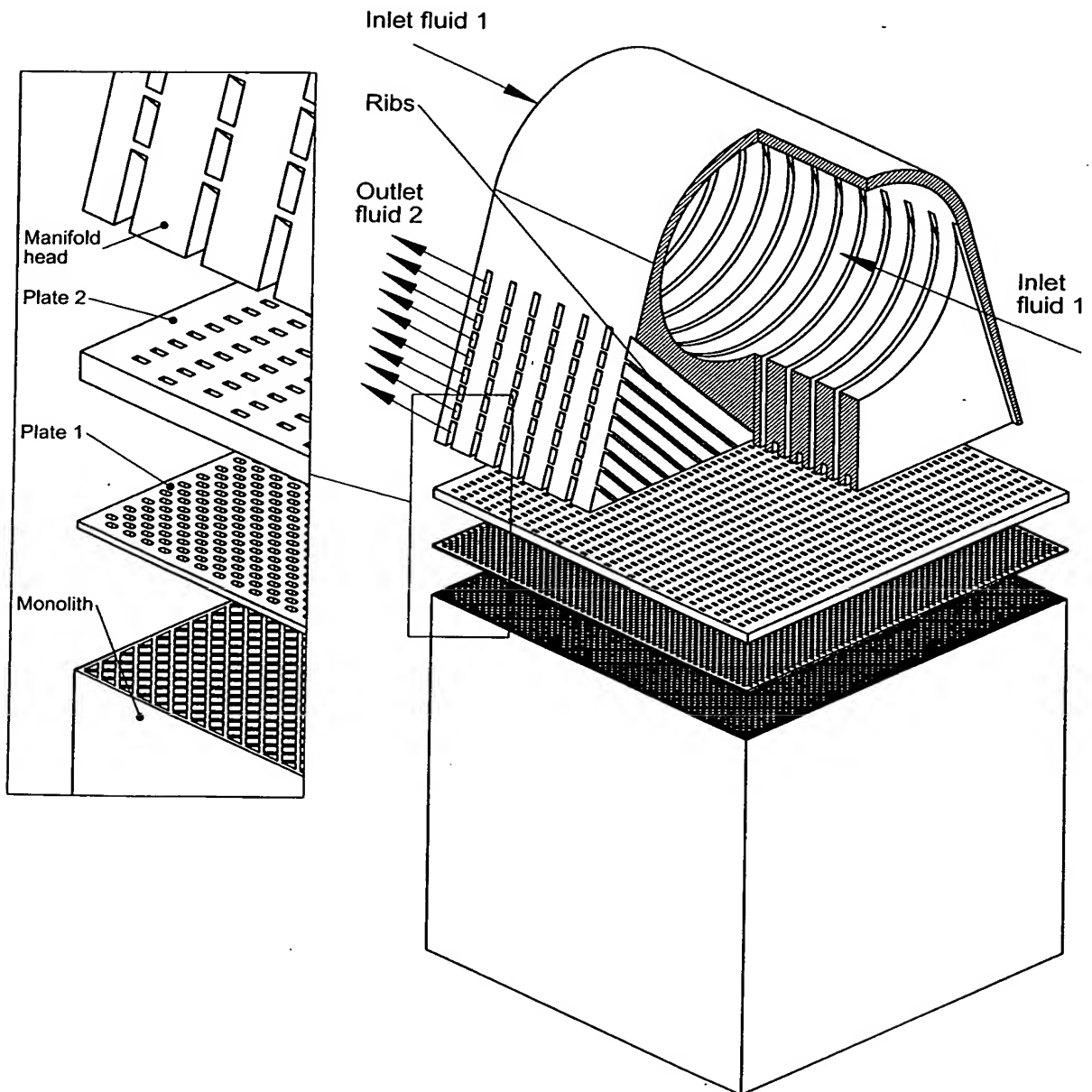


Fig. 16

**Fig. 17**

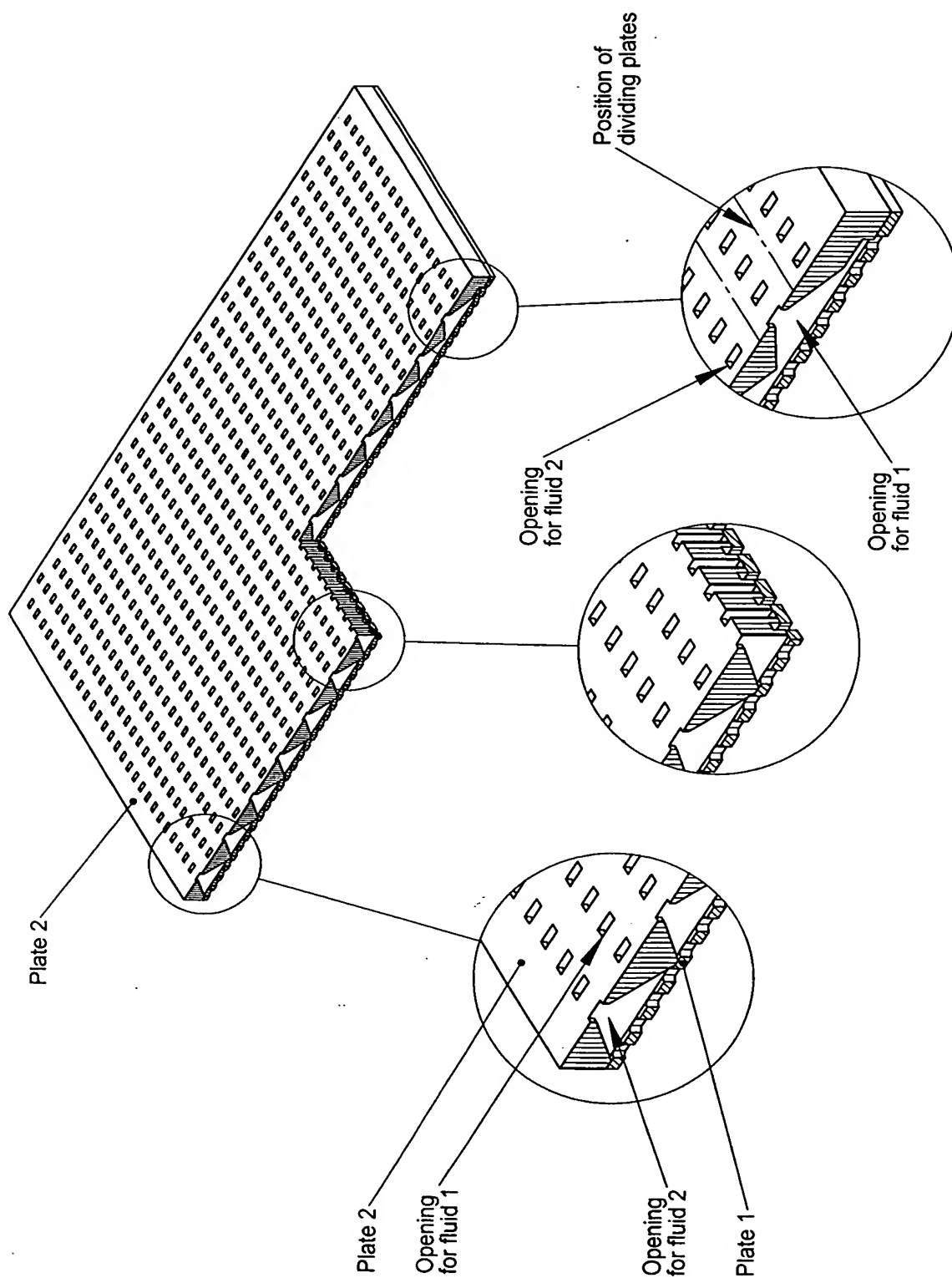


Figure 18